THESIS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

Black Liquor Gasification-Based Biorefineries – Determining Factors for Economic Performance and CO_2 Emission Balances

KARIN PETTERSSON

Heat and Power Technology
Department of Energy and Environment
CHALMERS UNIVERSITY OF TECHNOLOGY

Göteborg, Sweden 2011

Black Liquor Gasification-Based Biorefineries
– Determining Factors for Economic Performance and CO_2 Emission Balances
KARIN PETTERSSON
ISBN 978-91-7385-537-2

Doktorsavhandlingar vid Chalmers tekniska högskola
Ny serie nr 3218
ISSN 0346-718X

Publication 2011:1
Heat and Power Technology
Department of Energy and Environment
Chalmers University of Technology, Göteborg
ISSN 1404-7098

Chalmers University of Technology
SE-412 96 Göteborg
Sweden
Telephone + 46(0)31-772 1000

Printed by Chalmers Reproservice
Göteborg, Sweden, 2011

This thesis is based on work conducted within the interdisciplinary graduate school Energy Systems. The national Energy Systems Programme aims at creating competence in solving complex energy problems by combining technical and social sciences. The research programme analyses processes for the conversion, transmission and utilisation of energy, combined together in order to fulfil specific needs.

PROGRAM
ENERGISYSTEM
Forskning och forskarskola

The research groups that participate in the Energy Systems Programme are the Department of Engineering Sciences at Uppsala University, the Division of Energy Systems at Linköping Institute of Technology, the Department of Technology and Social Change at Linköping University, the Division of Heat and Power Technology at Chalmers University of Technology in Göteborg as well as the Division of Energy Processes at the Royal Institute of Technology in Stockholm.

www.liu.se/energi

Black Liquor Gasification-Based Biorefineries
– Determining Factors for Economic Performance and CO_2 Emission Balances

KARIN PETTERSSON
Heat and Power Technology
Department of Energy and Environment
Chalmers University of Technology

ABSTRACT

Biorefineries constitute an attractive future development option for the pulp and paper industry, allowing mills to produce not only pulp or paper but also other value-added products. Black liquor gasification (BLG) is currently being developed as an alternative technology for energy and chemical recovery in kraft pulp and paper mills. The technology enables the mill to increase the internal electricity generation or produce chemicals such as motor fuels. This thesis investigates the influence of different factors, including choice of product, type of mill, alternative investments, opportunities for carbon capture and storage (CCS) and future energy market conditions, on the economic performance and CO_2 emission balances for BLG-based biorefinery concepts.

Implementation of biorefinery concepts such as BLG with electricity production in future market pulp mills can be achieved without making the mill dependent on external wood fuel. However, implementation in integrated pulp and paper mills requires external wood fuel and reduces the amount of wood fuel available for other applications, thereby increasing the CO_2 emissions from those applications.

The results show that BLG with motor fuel production could be profitable for both market and integrated mills, whereas BLG with electricity generation is primarily an attractive option for market mills. For mills that operate with conventional recovery boiler technology, potentially profitable biorefinery concepts include lignin extraction or motor fuel production from gasified wood fuel. Few of the biorefinery concepts investigated in this work achieve a significant reduction of CO_2 emissions, especially for integrated mills. However, if commercially available, CCS could contribute to significant CO_2 emissions reduction and enhanced profitability for future energy market conditions characterized by a high CO_2 emissions charge, for both combustion- and gasification-based biorefinery concepts. Steam-saving measures could also significantly improve the economic performance as well as the CO_2 emission balances, especially for biorefinery concepts that use external wood fuel. The results also show that even if the recovery boiler has not reached the end of its technical lifetime, it could nevertheless be attractive for mills to consider investment in a smaller BLG plant.

Keywords: black liquor gasification, kraft pulp and paper mill, biorefineries, second generation biofuels, heat integration, system expansion, global CO_2 emissions, energy market scenarios

To my parents

List of appended papers

This thesis is based on the following papers, referred to by Roman numerals in the text:

I. Wetterlund E, Pettersson K and Magnusson M. Implications of system expansion for the assessment of well-to-wheel CO_2 emissions from biomass-based transportation. International Journal of Energy Research 2010;34(13):1136-1154.

II. Pettersson K and Harvey S. CO_2 emission balances for different black liquor gasification biorefinery concepts for production of electricity or second-generation liquid biofuels. Energy 2010;35(2):1101-1106.

III. Pettersson K and Harvey S. Economic Assessment of DME Production via Black Liquor Gasification Considering Different Future Energy Market Conditions and Mill Steam Balances. In Proceedings of International Chemical Recovery Conference, Williamsburg, Virginia, USA, March 29 – April 1, 2010;2:187-195.

IV. Pettersson K and Harvey S. The effect of increased heat integration on the cost for producing DME via black liquor gasification. In Proceedings of 22nd International Conference on Efficiency, Cost, Optimization, Simulation and Environmental Impact of Energy Systems (ECOS), Foz do Iguaçu, Paraná, Brazil, August 31 – September 3, 2009;1503-1512.

V. Pettersson K and Harvey S. Comparison of black liquor gasification with other pulping biorefinery concepts – Systems analysis of economic performance and CO_2 emissions. Submitted to Energy.

VI. Jönsson J, Pettersson K, Berntsson T and Harvey S. Comparison of options for utilization of excess steam and debottlenecking the recovery boiler at kraft pulp mills – Systems analysis of economic performance and CO_2 emissions. Submitted to International Journal of Energy Research.

Co-authorship statement

Pettersson is the main author of Papers II-V. Paper I is a joint effort of Wetterlund, Pettersson and Magnusson. Wetterlund was responsible for the input data regarding the plant configurations based on solid biomass gasification, Pettersson was responsible for the input data regarding the plant configuration based on black liquor gasification and Magnusson was responsible for the input data regarding the ethanol production plant. Paper VI is a joint effort of Pettersson and Jönsson. Pettersson was responsible for the calculations regarding black liquor gasification with downstream production of electricity and motor fuels, whereas Jönsson conducted system modelling and optimization in the energy system modelling tool reMIND. Professor Simon Harvey supervised the work in all papers and Professor Thore Berntsson supervised the work in Paper VI.

Related publications not included in this thesis

Fallde M, Flink M, Lindfeldt E, Pettersson K, Wetterlund E. Bakom drivmedelstanken – Perspektiv på svenska biodrivmedelssatsningar. Arbetsnotat. Program Energisystem, Linköping, Sweden, 2007. (*In Swedish*)

Flink M, Pettersson K, Wetterlund E. Comparing new Swedish concepts for production of second generation biofuels – evaluating CO_2 emissions using a system approach. In Proceedings of SETAC Europe 14th LCA Case Studies Symposium, Göteborg, Sweden, December 3-4, 2007;99-102.

Fornell R, Pettersson K, Berntsson T. Preliminary Design and Energy Efficiency Analysis of a Kraft Pulp Mill Converted to a Biorefinery Producing Ethanol and DME from Softwood. PRES 2010, 13th International Conference on Process Integration, Modelling and Optimisation for Energy Saving and Pollution Reduction, Chemical Engineering Transactions 2010;21:1147-1152.

Wetterlund E, Pettersson K, Harvey S. Systems analysis of integrating biomass gasification with pulp and paper production – Effects on economic performance, CO_2 emissions and energy use. Energy 2011;36(2):932-941.

Table of Contents

1 Introduction

With increased concern for climate change and security of supply, there is significant interest in replacing fossil resources with biomass feedstock for energy and material purposes. The EU has set targets for 20% reduction of greenhouse gas emissions, 20% share of renewables in the EU energy mix and 20% improvement of energy efficiency by 2020 (see for example European Commission, 2010). Furthermore, the EU has set a goal of 10% for the share of renewable energy for the transport sector for the year 2020 (European Union, 2009). Increased use of biomass could play an important role in reaching several of these goals. However, biomass is a limited resource which makes efficient resource utilization essential.

The pulp and paper industry is the sixth largest industrial energy user in Europe and it is a major user of biomass (CEPI, 2009). Through implementation of energy efficiency measures and increased delivery of energy and material products, the pulp and paper industry could make a significant contribution to achieving the goals set up within the EU.

Black liquor is formed during production of kraft pulp, which is the dominating method for chemical pulp production. In a conventional kraft pulp mill, black liquor is fired in a recovery boiler in order to recover energy, in the form of electricity and process utility steam, and pulping chemicals. Black liquor gasification (BLG) is currently being developed as an alternative technology for energy and chemical recovery. The major developer of BLG technology is the Swedish company Chemrec, and several development plants have been built or are planned in Sweden. In the gasification process the main fraction of the organic content in the black liquor is converted to a synthesis gas (syngas) and the pulping chemicals are recovered and returned to the pulping process, as for the recovery boiler process. The syngas can be used as a feedstock for production of renewable motor fuels (also referred to as biofuels) such as DME (dimethyl ether), methanol, FTD (Fisher-Tropsch diesel) or hydrogen. Such concepts are often referred to generically as black liquor gasification with motor fuel production, BLGMF. Alternatively, the syngas can be used as a fuel for electricity generation in a combined cycle cogeneration unit (a concept referred to as black liquor gasification combined cycle, BLGCC).

For the assessment of black liquor gasification, or other technology solutions for the pulp and paper industry, it is indispensable to have a reference case (alternative investment) to compare with. In order to limit the scope of studies and be able to focus on key novel aspects of the studied technology, it is natural to relate to a reference case based on some form of "business as usual", where proven technology solutions are assumed. However, in order to draw more general conclusions, it is important to highlight how competitive the technology is compared to other technologies under development. Furthermore, it is important to illustrate how the applicability and performance of competing technologies vary for different types of mills with different steam requirements.

BLG is one of several biorefinery options for the kraft pulp industry, enabling production of value-added products such as electricity, district heating, biofuels or lignin in addition to pulp. With increased energy and raw material (pulp wood) prices, investment in biorefineries is a possible way for the industry to remain competitive. Some mills, especially energy-efficient market kraft pulp mills, have the possibility to become major net exporters of electricity or lignin without purchasing external wood fuel. However, in order for integrated pulp and paper mills, even those with a high degree of energy efficiency, to become major exporters of lignin or for any type of mill to become a major exporter of biofuels, external wood fuel is required. In such cases the usage of biomass should be compared with other possible ways to use the biomass resource. Increasing the degree of heat integration could decrease the need for external process heating and thereby decrease the need for external wood fuel.

If the technology becomes commercially available, implementation of carbon capture and storage (CCS) could significantly influence both the climate impact and economic performance of the studied systems. In black liquor gasification, and other gasification processes, relatively large amounts of CO_2 could be captured at relatively low costs. Large amounts of CO_2 could obviously be separated from the flue gases of the recovery boiler, or other mill power boilers. However, the separation costs are generally significantly higher compared to implementation in the gasification processes.

In order for mills to consider implementation of full-scale BLG plants, the recovery boiler has to be close to the end of its technical lifetime. However, mills with a steam surplus, or mills planning to increase their production capacity (assuming that the recovery boiler is running at maximum capacity), could consider investment in a smaller BLG plant as a way to take advantage of a potential steam surplus or to achieve debottlenecking of the recovery boiler.

The focus on the transport sector's high oil dependence and climate impact has resulted in black liquor gasification being considered primarily as an option for production of biofuels in recent years. The technology is included in several studies comparing climate and economic benefits of alternative ways to produce motor fuels. However, there is a tendency to present black liquor gasification without consideration of the special implementation characteristics of each specific case. Few studies discuss how other characteristics, such as integration with another type of mill, would affect the results.

Estimating the climate impact and economic performance of possible future technologies is not straightforward. Uncertainties about, for example, future energy prices and policy instruments make the results highly variable. Furthermore, when it comes to estimation of climate impact, a number of different approaches can be considered, with significant variation of results.

1.1 Aim

The aim of this thesis is to illustrate and analyse the consequences for the energy system of integrating a black liquor gasification plant with subsequent production of biofuels or electricity. Different types of mills are considered, as well as various possible future scenarios for energy prices, policy instruments and other parameters that affect the economic performance and climate benefits of the integrated system. The influence of the following factors on the economic performance and CO_2 emission balances of black liquor gasification-based biorefinery concepts is discussed:

- Choice of product from gasified black liquor (biofuels or electricity)
- Mill steam requirements
- Mill investment requirements (recovery boiler replacement or capacity boosting)
- Alternative investments for the mill (recovery boiler-based options)
- Choice of technology for balancing a steam deficit/surplus at the mill
- The opportunities for CCS implementation at the mill
- Degree of heat integration
- Level of investment costs
- Methodology for evaluation of CO_2 emission balances
- Future energy market conditions (including policy instruments)

These factors will be further described and discussed in Chapter 4.

1.2 Papers

This thesis is based on six papers, which can be found in the end of the thesis. Below, the papers are briefly presented. Figure 1 illustrates the papers and which of the studied factors are included in which paper. From the figure we can see that all studied factors are included if Paper I and Papers IV-VI are considered. Paper V includes all the factors that are included in Papers II and III and investigates both the influence on economic performance and CO_2 emissions. Therefore, in order to limit the size of this thesis, results from Papers II and III are not explicitly discussed in the overview essay.

Paper I investigates the impact of expanding the system boundary to include the systems surrounding a biomass conversion system, when evaluating CO_2 emissions balances for different biomass-based transportation alternatives. In addition, the consequences of failing to expand the system boundary in an appropriate way are shown. Four cases of biomass-based transportation, including DME produced via BLG, are used to illustrate the system expansion method.

Paper II investigates how the CO_2 emission balances for different BLG concepts vary, depending on assumptions about future energy market conditions and mill steam requirements. Different final products are considered, including DME, methanol, FTD and electricity.

Paper III investigates how the economic performance of DME production via BLG depends on the mill steam demand level and future energy market conditions.

Paper IV shows the effect of increased heat integration on the economic performance and CO_2 emission balances of DME production via BLG for different future energy market conditions (calculations of CO_2 emission balances have been added and the economic calculations have been updated compared to the appended paper).

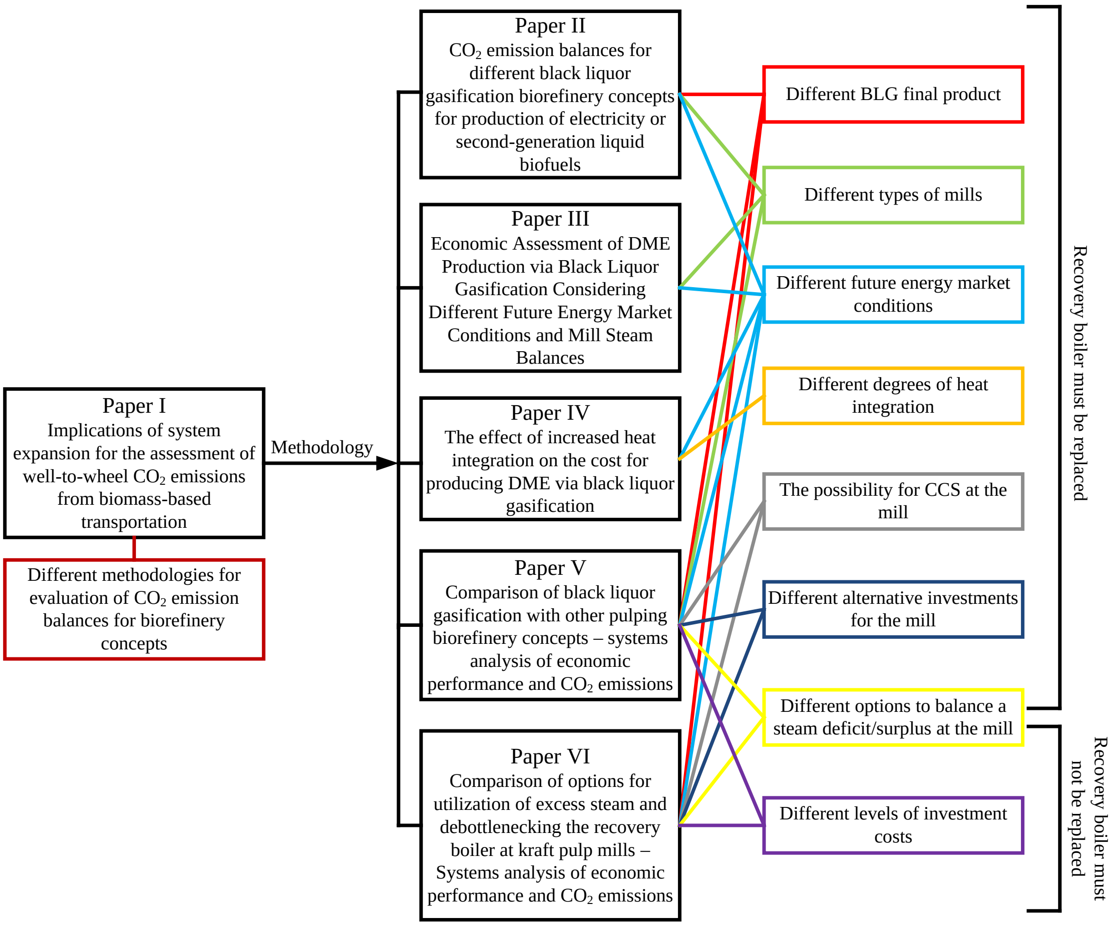

Figure 1. *Illustration of the papers included in this thesis and which of the studied factors they include.*

Paper V compares BLG with recovery boiler-based biorefinery concepts from economic and climatic point of views. The economic performance and CO_2 emission balances are calculated for different future energy market conditions. The study includes different types of mills, with different steam requirements, for which the applicability and performance of various biorefinery concepts vary. The possibilities for CCS in both combustion- and gasification-based systems are included.

Paper VI compares different technologies, including BLG, for utilization of excess steam and debottlenecking the recovery boiler at kraft pulp mills. The studied technologies are compared with respect to annual net profit and global CO_2 emissions for different future energy market conditions. In this paper, as a contrast to Papers I-V, the recovery boiler is not in need of replacement.

1.3 Thesis outline

Chapter 2 presents an overview of related studies investigating the consequences of implementation of BLG in kraft pulp and paper mills. Related studies regarding other pulping biorefinery concepts, heat integration opportunities in pulp and paper mills, and methodological issues are presented in Chapter 4.

Chapter 3 describes conventional kraft pulp production and the consequences of replacing the recovery boiler by a BLGCC or BLGMF plant. Thereafter follow more detailed descriptions of possible future BLGCC and BLGMF plants. After that, an overview of the historical development of the technology is provided. Finally, a short presentation of the availability of black liquor in the world is given.

Chapter 4 describes and discusses the factors influencing the economic performance and CO_2 emissions balances of BLG systems which are studied in this thesis. The factors include both more technical issues, such as which final products and types of mill are considered, and methodological issues regarding evaluation of CO_2 emission balances and economic performance of future biomass-based conversion systems. An overview of the extent to which the different parameters have been included in previous studies as well as in the different papers in this thesis is also presented.

Chapter 5 presents the methodology used for calculation of economic performance and CO_2 emission balances, including the energy market scenarios used. Chapter 6 presents a summary of the papers included in this thesis. Chapters 7-9 include a discussion of the results, conclusions and suggestions for further work.

2 Related studies

This chapter presents an overview of related studies investigating the consequences of implementation of the Chemrec black liquor gasification technology in kraft pulp and paper mills. An overview of related studies regarding other pulping biorefinery concepts, heat integration opportunities in pulp and paper mills, and methodological issues is presented in Chapter 4.

As in this thesis, previous studies have mostly focused on BLG with downstream production of electricity or motor fuels. Studies of the consequences of implementation of BLGCC technology have been performed by, for example, Berglin (1996), Näsholm and Westermark (1997), Maunsbach et al. (2001), Eriksson (2001), Möllersten (2002), KAM (2003), Larson et al. (2003), Modig (2005) and Joelsson and Gustavsson (2008). Some studies are mainly focused on detailed process modelling and energy analysis (for example Berglin, 1996) whereas other studies mainly focus on environmental and/or economic analysis (for example Joelsson and Gustavsson, 2008). There are also more extensive studies that include detailed process modelling as well as economic and environmental analysis at both the mill and national/regional levels (for example Larson et al., 2003).

The consequences of implementation of BLGMF technology have been studied by, for example, Isaksson (2000), Möllersten (2002), Ekbom et al. (2003; 2005), Larson et al. (2006; 2008; 2009), Andersson (2007) and Joelsson and Gustavsson (2008). As for studies related to BLGCC technology, there are both studies that mainly focus on a specific system level (for example Joelsson and Gustavsson, 2008), and studies that include both detailed process modeling and economic and/or environmental analysis (for example Ekbom et al., 2003; Ekbom et al., 2005; Larson et al., 2006).

The studies related to BLGCC technology date mainly from the mid-1990s to the mid-2000s, whereas the studies related to BLGMF technology have mostly been conducted during the past decade. BLG is also included in studies that compare several different options for using biomass or compare several different routes for production of motor fuels (see for example Edwards et al., 2007; Gustavsson et al., 2007; Renew, 2008).

Berglin (1996), Berglin and Berntsson (1998) and Berglin et al. (1999) studied how the introduction of BLG technology with electricity generation affects the mass and energy balances for different types of mills. The opportunities for energy savings through increased integration between the gasification plant and the other mill processes were investigated. Models for the gasifier and other parts of the systems such as the gas turbine cycle were developed in order to study the influence of a number of different parameters (gasification agent, gasification temperature, dry solid content of the black liquor, etc.). The possibilities for increased electricity generation with BLG technology, compared to recovery boilers with different steam data levels, were studied. The results showed that both market and integrated pulp mills have the possibility to become net exporters of electricity if black liquor gasification with electricity generation is implemented. For integrated mills this generally requires additional external fuel.

The potential for increased electricity generation with BLG technology was also investigated by Näsholm and Westermark (1997). Their study includes different cogeneration technologies based on black liquor gasification, including BLGCC. Maunsbach et al. (2001) investigated the integration of combined cycles and advanced gas turbines based on both black liquor and solid biomass gasification. Both studies conclude that black liquor gasification offers a significant potential for increased electricity generation in the pulp and paper industry compared to conventional recovery boiler-based technology.

Eriksson (2001) showed the importance of correctly considering the surrounding system when selecting mill powerhouse technology and CHP (combined heat and power) plant configuration if the system as a whole (mill system and surrounding system) should maximize the electricity production from a given fuel resource. BLG with different gas turbine cycles, as well as cases that only produce heat from the syngas and recovery boilers with different steam data, were considered for integration with different types of mills. The results showed that black liquor gasification technology is attractive in all cases compared to recovery boiler technology.

In KAM (2003) the mass and energy balances for integration of a BLGCC plant to a market pulp mill were calculated. By estimating the incremental investment and running cost, compared to a new recovery boiler, the cost for the extra electricity that is produced in the BLGCC system, compared to the recovery boiler system, was estimated.

Larson et al. (2003) performed an assessment of the prospective energy, environmental and economic performance of BLGCC plants in integrated pulp and paper mills in the United States. Comparisons to recovery boiler technology with different steam data levels were included. It was found that BLGCC systems can provide energy, environmental (such as reduced CO_2 emissions) and economic benefits at the mill level, as well as at national and regional levels.

Modig (2005) analyzed the technical, economic and societal conditions for introducing BLG technology. The importance of mill steam demand level and recovery boiler steam data for the difference in total and electrical efficiency between recovery boiler- and

BLG-based CHP production options was shown. It was also shown that these factors, together with other factors such as the assumed capital cost, significantly influence the economic performance of BLG with electricity production and determine whether the technology is profitable or not. Opportunities and hurdles for the introduction of black liquor gasification were also discussed. The study concludes that there are many benefits with gasification, such as the possibility to use the syngas for production of a variety of products and the possibility to easily capture and store CO_2. The hurdles include high investment costs, low reliability and risks associated with an unproven technology.

Isaksson (2000) examined the greenhouse gas (GHG) reduction potential if black liquor gasification with co-production of biofuels (hydrogen, methanol, FTD) and electricity (and process steam) is implemented in the pulp industry. The results showed that hydrogen is the only biofuel that has higher CO_2 reduction potential compared to the mill with a conventional recovery system or a mill implementing BLGCC technology.

Möllersten (2002) investigated opportunities for CO_2 emissions reduction in the pulp and paper industry through, for example, implementation of black liquor gasification technology. The results show that a BLGCC plant can contribute to large CO_2 emission reductions and that the reduction potential can be significantly increased, at a relatively low additional cost, if CCS is considered. Alternatively, black liquor gasification with co-production of methanol and electricity and inclusion of CCS could contribute to large CO_2 emission reductions. The influence of the assumptions regarding the electricity system in the fossil fuel-based reference energy system for the assessment of global CO_2 emissions reduction potential is shown.

Berglin et al. (2003) calculated the mass and energy balances for the integration of BLGCC and BLGMF/methanol plants to market and integrated mills. Ekbom et al. (2003; 2005) performed an assessment of the energy and economic performance of BLGMF plants producing methanol, DME or FTD in market pulp mills. The study concludes that there are substantial economic incentives for making investments in BLGMF plants. The total efficiency and profitability are greater for DME and methanol than for FTD. The study was performed for Swedish conditions.

Larson et al. (2006; 2008; 2009) performed an assessment of the prospective energy, environmental and economic performance of gasification-based biorefinery options, including both gasification of black liquor and solid biomass, for production of different biofuels and electricity in integrated pulp and paper mills in the United States. DME, FT liquids, mixed alcohols and electricity were considered as main products. The study concludes that, once commercialized, gasification-based biorefinery technologies offer the potential for attractive investment returns as well as energy and environmental benefits for the country. Whether electricity or biofuels are most profitable is dependent on future energy price assumptions. The potential to reduce CO_2 emissions, per unit of biomass used, is higher for electricity production than for biofuel production.

Andersson (2007) compared the CO_2 emissions reduction potential of BLG with hydrogen, methanol or electricity production in a market pulp mill under different future energy market scenarios. The results show that hydrogen has a significantly higher potential for reduction of CO_2 emissions compared to methanol in all studied scenarios. If CCS is considered, the difference in potential CO_2 emissions reduction between the biofuels increases further. Whether hydrogen or electricity achieves the highest reduction potential is dependent on the assumptions regarding the electricity and transport system in the fossil fuel-based reference energy system. It is also shown that the production costs are lower, and the CO_2 emissions reduction potential are higher, for hydrogen production via gasification of black liquor, with use of external wood fuel to cover the mill steam deficit, compared to using the same amount of wood fuel for hydrogen production via gasification of the same amount of wood fuel integrated with a natural gas combined cycle (NGCC) CHP or district heating system.
.
Joelsson and Gustavsson (2008) studied the potential for reduction of CO_2 emissions and oil usage associated with implementation of BLGCC and BLGMF technology in market and integrated mills. The oil reduction potential is obviously higher for BLGMF plants compared to BLGCC plants (which can lead to increased oil use), whereas the potential to reduce global CO_2 emissions is dependent on the assumptions regarding the electricity system in the fossil fuel-based reference energy system. However, the potential to reduce CO_2 emissions, per unit of biomass used, is higher for BLGCC plants regardless of assumptions regarding the electricity system. If stand-alone production of electricity and transportation fuels from biomass is included to balance the systems compared, so that they achieve the same CO_2 emissions and oil use reductions, it is more efficient to implement BLG with motor fuel production and stand-alone electricity production, than to implement BLG with electricity production and stand-alone production of motor fuels. Joelsson et al. (2009) conclude that it is more efficient to produce motor fuels integrated with pulp and paper mills than in stand-alone plants. The efficiency for motor fuel production via black liquor gasification is higher in market pulp mills than in integrated pulp and paper mills, due to low total energy efficiency for the market pulp mill reference case. Furthermore, it is concluded that to use the biomass for replacement of coal, instead of producing motor fuels, achieves higher reduction of CO_2 emissions.

McKeough and Kurkela (2008) compared the estimated performance and costs of methanol production via solid biomass integrated with a pulp and paper mill with methanol production via black liquor gasification (naturally integrated with a pulp mill). The thermal efficiency was shown to be similar in both cases, whereas black liquor gasification was found to be somewhat more economical.

In several of the mentioned studies it is stressed that projected commercial ("Nth plant") performance and costs (if included) are assumed. All the studies consider a full substitution of the recovery boiler. However, Berglin et al. (2003) considered a case where part of the black liquor is gasified and part is combusted in the recovery boiler. In this way no wood fuel was needed in order to satisfy the mill steam demand. Berglin and Andersson (2001) investigated different possibilities for additional black liquor processing requirements, resulting from a production capacity increase, in a mill where the recovery boiler already was running at maximum capacity and could not be rebuilt. They concluded that a black liquor gasifier using the product gas for steam generation yields a better economic return than investing in a new recovery boiler.

Edwards et al. (2007) performed a study where energy and GHG emission balances were calculated for a number of different motor fuels and powertrains, both conventional and innovative. The whole chain from fuel resource to performed transportation work was considered. Motor fuel production via BLG was found to have lower energy and GHG emissions compared to production via solid biomass gasification. Production of DME was found to have better energy and GHG emissions results compared to other liquid fuels such as FTD.

Gustavsson et al. (2007) considered both BLGCC and BLGMF technologies, together with several other options, for using a limited biomass resource for reduction of global CO_2 emissions and oil use. Electricity and motor fuel production via BLG were found to be attractive options for CO_2 emissions and oil use reduction respectively. Both BLGMF and BLGCC technologies are of interest if a strategy where both these objectives are to be fulfilled simultaneously is adopted.

In Renew (2008) a technical, economic and environmental assessment of different biofuels based on a lignocellulosic feedstock was performed. A number of different production routes, mainly gasification-based, were considered for production of mainly FTD or DME. Amongst the studied production routes, BLG with DME production was found to have the lowest biofuel production cost and the lowest global warming potential.

3 Black liquor gasification

Pulp is produced in two main ways: by chemical or mechanical separation of the cellulose fibres. The kraft (sulphate) process is the dominating method for chemical pulp production. In Sweden, for example, it accounts for more than 90% of the chemical pulp production (Swedish Forest Industries, 2009). The rest of the production is mainly from the sulphite process. Black liquor is formed during production of kraft pulp. In the sulphite process, a similar spent cooking liquor is formed which also could be a feedstock for gasification. Furthermore, there is ongoing research about the possibility to convert kraft pulp mills for production of ethanol, whereby the cellulose is used for ethanol production instead of pulp production (Fornell et al., 2010). A cooking liquor, similar to black liquor, is also formed in such a process, which could be gasified.

This thesis is about black liquor gasification coupled to the kraft pulp process. More specifically it concerns the pressurized, high-temperature (950-1000°C), oxygen-blown, entrained-flow black liquor gasification technology developed by Chemrec in Piteå, Sweden. In the USA and Canada another gasification technology has been developed during the past decades. The process is based on low-temperature (~600°C) gasification with steam in a bubbling fluidized bed, and has been developed by ThermoChem Recovery International (TRI) (Berglin, 1996; Larson et al., 2003; Modig, 2005; Larson et al., 2006; Lindström et al., 2007). The technology has however only been tested on liquor from the soda process (a third alternative for chemical pulp production besides the kraft and sulphite process), not on sulphur-rich black liquor from the kraft process, and severe doubts about the technical feasibility of the TRI process have been raised (Modig, 2005). Currently, one plant is in operation at a mill in Trenton, Canada (TRI, 2011). In, for example, Berglin (1996) and Larson et al. (2003) both high-temperature (Chemrec) and low-temperature (TRI) gasification are considered.

This chapter will first describe conventional kraft pulp production (see for example Biermann, 1996). Thereafter, the consequences of replacing the recovery boiler with a BLGCC or BLGMF plant are described. Next follow more detailed descriptions of possible future BLGCC and BLGMF plants. An overview of the development of the technology is then provided, including both a historical overview and the current situation. Finally, a short presentation of the availability of black liquor will be given.

3.1 Conventional kraft pulp production

After the pulp wood has been debarked and cut into wood chips, it is added to the digester where it is mixed with cooking liquor, known as white liquor, containing the cooking chemicals (NaOH and Na_2S) and water. Cellulose fibres in the wood chips are separated from the lignin, which acts as glue between the fibres, when the lignin reacts with the chemicals in the white liquor. The chemicals and lignin form a liquor called black liquor (also contains other substances, mainly hemicelluloses). The fibres are separated from the black liquor in a washing step and are then screened and bleached before pulp is obtained. The bleached pulp is either dried and transported to a paper mill (this is called a market pulp mill), or processed further to paper at the mill (called an integrated pulp and paper mill).

The black liquor, which contains large amounts of water (only 15-20% dry solid content), is evaporated to a dry solid content of 70-80% before it is burned in a special boiler, called a recovery boiler. In the recovery boiler, energy is released in the form of exhaust gases used to produce steam. The remainder of the liquor can be found at the bottom of the boiler in the form of a smelt. The smelt is dissolved to form green liquor, which is sent to the chemical preparation where white liquor for the digester is produced. Thus, the recovery boiler functions both as an energy and chemical recovery unit.

In the white liquor preparation, CaO is used to regenerate NaOH. CaO is dissolved in water to form $Ca(OH)_2$ which reacts with Na_2CO_3 and forms NaOH and $CaCO_3$ (lime). CaO is then regenerated in the lime kiln where $CaCO_3$ is heated up and forms CaO and CO_2. Fuel oil is the most commonly used fuel in lime kilns today. However, as the pulp and paper industry tries to decrease the usage of fossil fuels, some mills use other fuels in the lime kiln, such as fuel gas produced by gasifying falling bark (see for example Möllersten, 2002).

The high-pressure (HP) steam produced in the recovery boiler is used in a back-pressure steam turbine (ST). The steam is then used to satisfy the heating requirements in the pulping process, such as digesting, evaporation and drying. Normally two different pressure levels of process steam are used at a mill: low- and medium-pressure steam (LP and MP steam). In cases where the steam from the recovery boiler is not sufficient to satisfy the mill steam demand, an additional boiler is used to produce HP steam for the back-pressure turbine. Fuel for this boiler is often bark from the debarking of the logs, possibly supplemented by purchased bark or other wood fuels and/or fuel oil. A surplus of steam can also occur, that is, more steam is produced by the recovery boiler than is needed at the mill. This steam could for example be used to produce additional electricity in a condensing steam turbine. Figure 2 shows a simplified illustration of the main energy and material streams in a conventional kraft pulp mill with a recovery boiler.

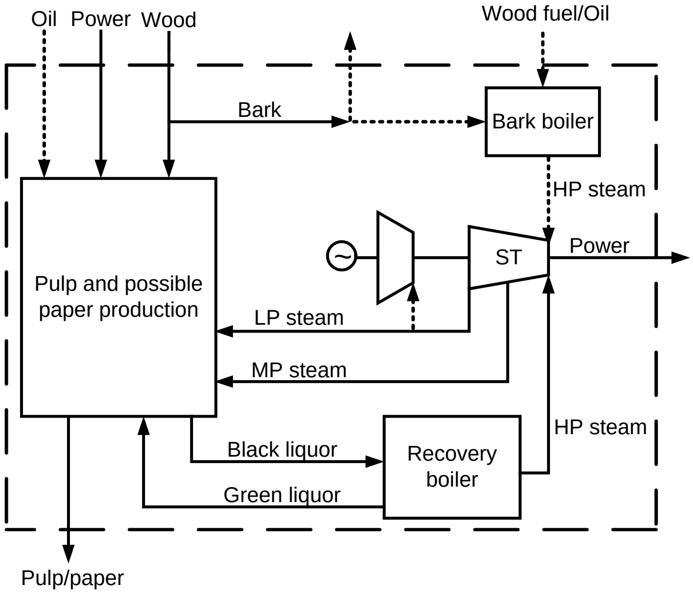

Figure 2. *Simplified illustration of the main energy and material streams at a conventional kraft pulp mill with a recovery boiler. Solid lines represent flows that are always relevant whereas dotted lines represent possible flows.*

A relatively common energy flow that is not included in Figure 2 is district heating. If located within reasonable distance from a district heating network, excess steam or heat from the mill could be used for production of district heat.

There is a lot of ongoing research and development connected with possible new technologies in the kraft pulp industry. Examples, besides gasification of black liquor, are extraction of lignin from the black liquor or hemicelluloses from the wood. Furthermore, for mills with a steam deficit, gasification of solid biomass with production of, for example, electricity or motor fuels and steam could be an alternative to the bark boiler for covering the steam deficit. More about this can be found in Sections 4.4 and 4.6.

3.2 Kraft pulp production with black liquor gasification

Replacing the recovery boiler with a black liquor gasification plant will change the mill's energy balance. Figure 3 shows a simplified illustration of possible main energy and material streams at a future kraft pulp mill with a BLGCC plant or a BLGMF plant.

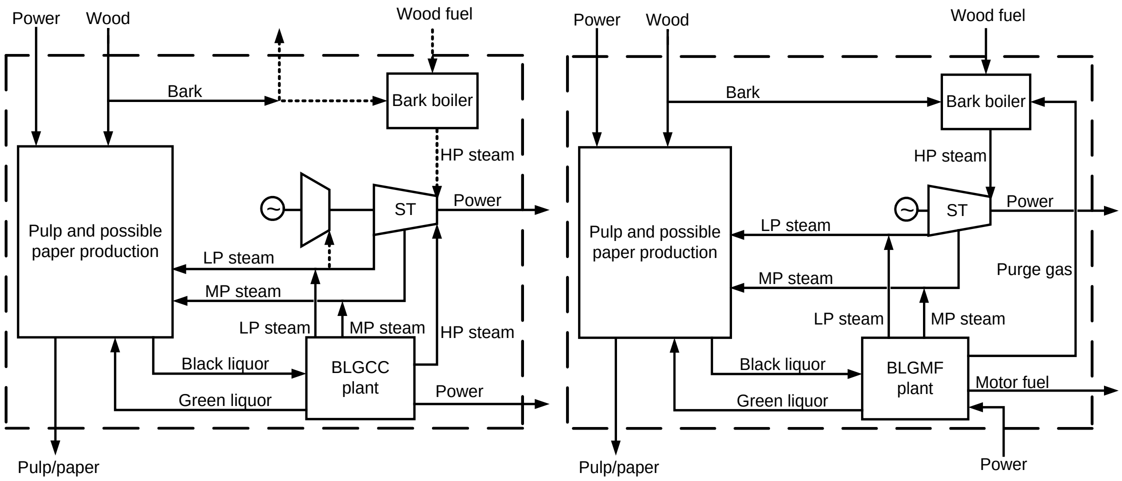

Figure 3. *Simplified illustration of possible main energy and material streams at a future kraft pulp mill with a BLGCC plant (with the steam turbine illustrated separately, left part of the diagram) or a BLGMF plant (right part of the diagram). Solid lines represent flows that could be relevant for all types of mills, whereas dotted lines represent flows that could be relevant for some types of mills.*

Black liquor is gasified with oxygen at high pressure in an entrained flow gasifier. The produced gas is cooled and cleaned before it is sent to the gas turbine or to the motor fuel synthesis. The chemicals are recovered as green liquor and sent to the white liquor preparation, as for the recovery boiler case.

Excess heat at suitable temperatures from the BLGCC and BLGMF plants can be used to generate steam. Some steam is used internally, but there is a significant surplus that can be used in the mill processes. However, it should be noted that less steam is produced compared to the conventional recovery boiler powerhouse configuration, since either motor fuels or more electricity are produced in the case of BLG. If the steam surplus from the BLG plant cannot cover the mill steam demand, additional steam could be produced in a bark boiler. The fuel used for this purpose will in the future most likely be (apart from the falling bark) purchased wood fuel. As mentioned in the previous section, it could also be interesting to implement a biomass gasification plant instead of a bark boiler (see further Section 4.6).

In order for a kraft pulp mill with a BLGMF plant to avoid having a deficit of steam, the mill steam demand must be extremely low (see Paper III). Therefore, in Figure 3, the mill is illustrated as having a steam deficit. It is also assumed that future mills will not use fuel oil, but rather wood fuel, as fuel in the lime kiln (therefore no oil is purchased to the mill; see Figure 3). Purge gas, formed in the motor fuel synthesis in the BLGMF case, could be used as fuel in the bark boiler, as indicated in Figure 3, in the lime kiln or in a gas turbine. The mill's power generation will obviously decrease, and the power consumption will increase, if the recovery boiler is substituted with a BLGMF plant (supplemented by a bark boiler cogenerating electricity).

Almost all sulphur ends up in the green liquor in the case of recovery boiler operation, whereas some of the sulphur in the black liquor ends up in the gas in the form of H_2S in the case of black liquor gasification (Modig, 2005). If the H_2S is reabsorbed in the white liquor, there is no effect on the pulp yield and properties (some CO_2 is absorbed together with the H_2S). However, there is a potential to produce cooking liquors with different sulphide contents. This improves the yield of pulp and thus the same amount of pulp can be produced using less wood. This method, called polysulphide cooking, has been considered in connection with black liquor gasification in, for example, Isaksson (2000), KAM (2003) and Larsson et al. (2006). The lower sulphur content in the green liquor leads to a higher content of carbonate (Na_2CO_3), and consequently the white liquor preparation requires more CaO and the load of the lime kiln increases (KAM, 2003).

Since the gasifier unit has no smelt bath, there is no risk of explosion, contrary to a recovery boiler (Modig, 2005). The smell from the mill will also decrease because the presence of reducing sulphur substances decreases compared with recovery boiler operation, since the synthesis gas undergoes an efficient sulphur treatment (Ekbom et al., 2005). Furthermore, it is easier to capture the CO_2 in case of black liquor gasification (see Section 4.5).

Introduction of black liquor gasification will make the process more complicated, compared to conventional recovery boiler operation (KAM, 2003; Modig, 2005). The investment cost is significantly higher than for recovery boiler technology, especially for BLGMF plants. Furthermore, there are challenges when it comes to, for example, increased concentration of non-process elements such as potassium and chlorine that could lead to increased problems with deposits and corrosion, finding materials that can handle the more corrosive green liquor that is formed in the case of black liquor gasification, and achieving stable and continuous operation (see further Section 3.4) (KAM, 2003; ETC, 2011).

3.3 From black liquor to final product

Black liquor enters the entrained-flow gasifier together with oxygen and is gasified at approximately 25-35 bar and 950-1000°C (KAM, 2003; Ekbom et al., 2005; Larson et al., 2006; Landälv et al., 2010). In several studies, for example KAM (2003) and Ekbom et al. (2005), of future implementation of BLGMF and BLGCC plants, a black liquor dry solid content of 80% is assumed. The following description is based on Ekbom et al. (2005) and KAM (2003).

Gasification is a thermo-chemical process in which carbon compounds in solid or liquid phase undergo decomposition. The decomposition occurs with a stoichiometric oxygen deficit, which means that the chemical reactions cannot proceed to carbon dioxide and water as for combustion. A high-energy gas consisting mainly of H_2, CO, H_2O, CO_2 and some H_2S and CH_4 is formed. The gasifier is adiabatic, that is, heat for the endothermic gasification reactions is provided by the exothermic oxidation reactions (combustion reactions). In the black liquor gasifier, smelt droplets containing the cooking chemicals (the temperature is above the melting point of the chemicals) are also formed. A quench zone in which the synthesis gas and the smelt are cooled by injection and evaporation of condensate from the following gas cooling unit is located in the bottom of the reactor. The smelt droplets are solidified, precipitate and dissolve in weak liquor (diluted white liquor) at the bottom of the quench zone and green liquor is formed. The green liquor is then sent to the white liquor preparation. The drop-free gas is cooled further by contact with more condensate from the gas cooling unit. The gas from the quench is sent to the bottom of the gas cooling unit, which is designed as a counter-current condenser. As the gas flows upwards it is counter-currently washed by the formed condensate flowing downwards. The lower and warmest section of the gas cooling unit is used to generate steam, whereas the upper section can be used for example to preheat make-up boiler feed water (BFW). The final cooling is achieved by cooling water (CW). Figure 4 shows an illustration of the Chemrec gasifier and the counter-current gas cooler.

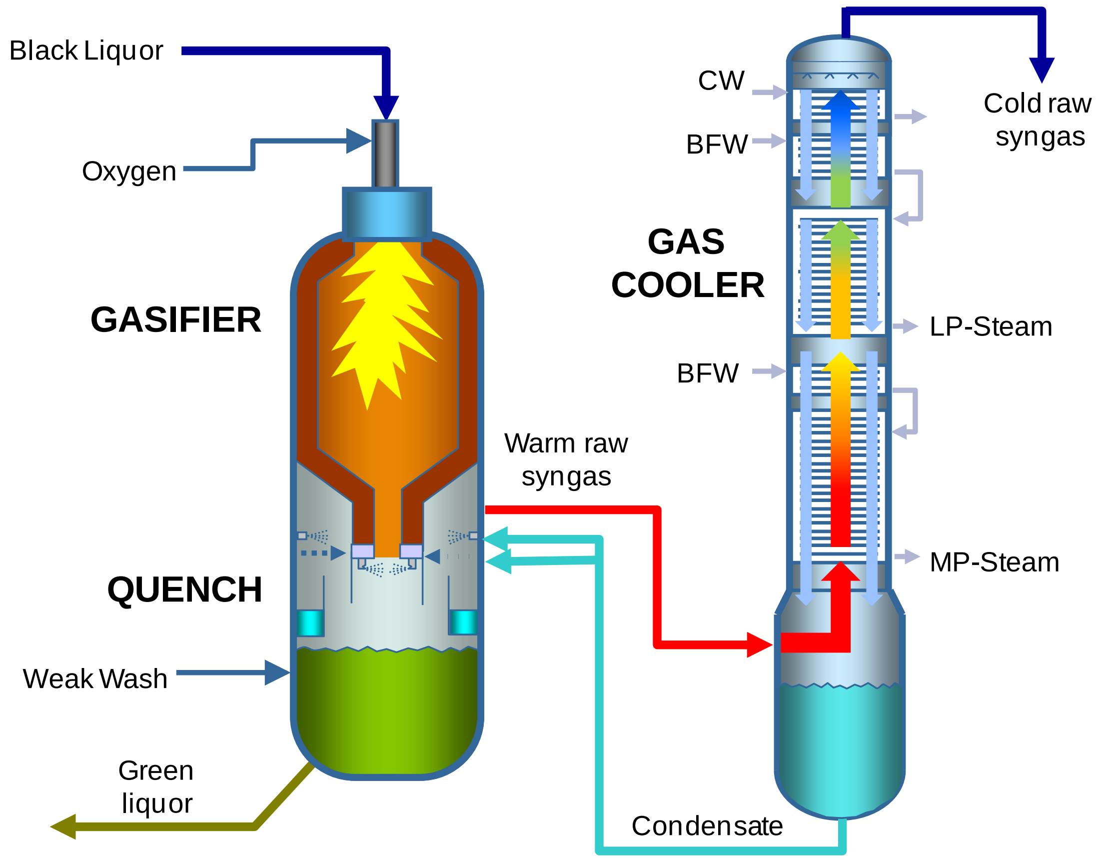

Figure 4. *The pressurized entrained-flow black liquor gasifier of Chemrec type and the counter-current gas cooler (based on Chemrec, 2005b).*

The design of the further cleaning and processing of the gas depends on whether the gas is used for electricity generation or for synthesis of motor fuels. In the case of electricity generation, the gas is cleaned from H_2S by, for example, absorption in nitrogen (Selexol unit). The cleaned gas is fired in a gas turbine (GT) for electricity generation. The exhaust gas then passes through a heat recovery steam generator (HRSG), generating HP steam for additional electricity generation in a steam turbine. The exhaust gas is also used to generate MP/LP steam and could, for example, preheat make-up boiler feed water. Figure 5 shows an illustration of the main energy and material flows in a BLGCC plant. Steam/heat is needed in order to regenerate the absorbent used for the gas cleaning and preheat the gas prior to combustion in the gas turbine cycle. Simple gas turbine cycles (no steam turbine is used; only steam directly for the process is produced in the HRSG) could also be used, as considered by for example Berglin (1996) and Eriksson (2001).

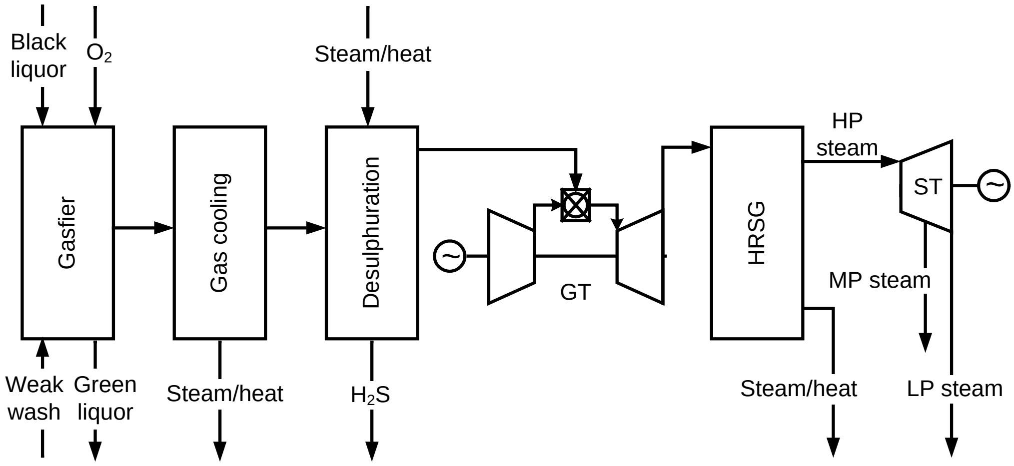

Figure 5. *Main energy and material flows in a BLGCC plant (electricity usage not included).*

Catalytic synthesis units for production of synthetic fuels require a high-purity syngas. Furthermore, the gas contains impurities such as tars that have to be completely removed. The gas is cleaned by using for example a Rectisol unit. First, a pre-wash unit is used to remove tars using methanol as solvent. Then H_2S and CO_2 (and some COS) are removed by absorption in methanol. The methanol is then regenerated and separated from the absorbed compounds. H_2S, CO_2 and COS are sent to a sulphur recovery unit. The gas has a H_2/CO ratio that is too low for synthesis of methanol, DME (if the DME is produced via methanol) and FTD. This is handled by using a water-gas shift reactor where CO is reacted with H_2O (steam is added) to produce H_2 and CO_2. This reaction is strongly exothermic and the shifted gas stream is cooled by raising steam. The gas is then sent to a second absorber in the Rectisol system for removal of the CO_2 formed in the water-gas shift reaction.

DME, which is considered as an example of a possible future motor fuel in this thesis (see Section 4.1), can be produced via synthesis of methanol. First, the cleaned syngas is compressed (from around 30 bar to 90-100 bar). Then DME is produced by reactions in three reactors. The reactions are strongly exothermic and the reactors are cooled by raising steam. Finally, the produced DME is purified in a distillation unit. Figure 6 illustrates the main energy and material flows in a BLGMF plant with production of DME. Steam/heat is needed in order to regenerate the absorbent used for removal of H_2S and CO_2 and for the DME distillation.

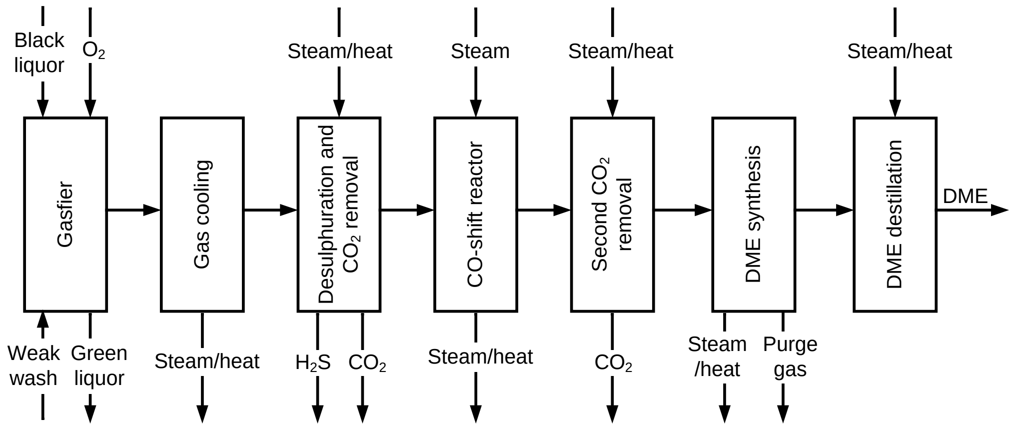

Figure 6. *Main energy and material flows in a BLGMF/DME plant (electricity usage not included).*

For a detailed description of production of DME and also methanol and FTD, see Ekbom et al. (2003) and Ekbom et al. (2005). Andersson (2007) presents a description of hydrogen production from gasified black liquor. The conversion efficiency from black liquor to fuel is almost the same for all these cases, approximately 50-55% (based on LHV). However, in the case of FTD production about two thirds are diesel and one third is another product, for example naphtha. The amount of excess steam/heat differs between the processes, and the usages of the different fuels require different degrees of changes in existing distribution infrastructure and of existing combustion engines (see Section 4.1 for more information about distribution and usage of different fuels).

Production of DME via methanol is a commercial process. There is ongoing development of direct synthesis of DME (see for example Larson, 2006). The required H_2/CO ratio then changes from 2 to 1, thereby avoiding the need for a CO-shift reactor and thus a second CO_2 removal step.

3.4 Development of the BLG technology – historical overview and current situation

3.4.1 Development of gasification technology

Combustion has been the totally dominant method to oxidize carbon-containing materials and thereby release heat, ever since people learned how to make a fire – and the absolute majority of bioenergy applications are based on combustion. Gasification is an alternative method for extraction of the stored chemical energy from biomass. Gasification technology has been known for over 100 years and was for example used during World War II to produce gas for vehicles from wood. Both solid and liquid fuels can be gasified, such as residual oils, waste, biomass and coal. Coal is currently the predominant feedstock for commercial gasification plants, whereas biomass so far represents a negligible share. Biomass gasification is still in the development phase. With the exception of less advanced applications, such as production of fuel gas, there are still no large-scale commercial biomass gasification plants, which there are for the other feedstocks mentioned. (Modig, 2005; Hellsmark, 2010)

The interest in biomass gasification has increased steadily during recent decades. The basic driving force for gasification of biomass is the ability to achieve a higher share of high-quality energy carriers, such as electricity and biofuels, from biomass compared to conventional biomass conversion technologies (Nyström et al., 2007). In the 1990s the focus was mainly on electricity generation, including replacement of conventional steam cycles with gas turbines in order to increase the electrical efficiency, which was also stimulated by the substantial development of gas turbines towards higher efficiencies and lower prices that occurred during this time (Berglin, 1996; Stevens, 2001). During the early 2000s, there was a clear increase of the interest in biomass gasification as a way to produce motor fuels and chemicals, mostly hydrogen for use in fuel cells (Stevens, 2001). That the focus has shifted to motor fuels can be seen as a reaction to the transport sector's climate impact, along with a desire to reduce dependence on fossil oil and concerns about political stability in a number of major oil-producing countries. Meanwhile, conventional combustion plants have been developed in order to achieve higher steam data and matching with more advanced steam cycles, driven by higher prices for electricity (Nyström et al., 2007). More advanced steam cycles and steam data have significantly decreased the electrical efficiency gap between combustion- and gasification-based power generation. However, since the electricity price is significantly higher today than in the 1990s, it is not given that the difference in electricity revenues between gasification- and combustion-based power generation has decreased. Currently, work is performed in many parts of the world to develop sustainable, profitable gasification processes for commercial production of fuels, electricity or other products from biomass (see for example Hellsmark (2010) for an overview of ongoing biomass gasification projects).

3.4.2 Alternatives to recovery boiler technology

The recovery boiler, which has been used for about 70 years, has struggled over the years with a series of technical problems such as corrosion, fouling, explosion hazards and high emission levels. This has resulted in many different ideas and suggestions for other technical solutions for energy and chemical recovery. Among these proposals a number are based on gasification instead of combustion. The problems of the recovery boiler have been greatly reduced over the years, so currently the main drivers for black liquor gasification, as well as for other gasification technologies, are opportunities for higher electrical efficiency and to produce other products such as motor fuels. (Modig, 2005) For an overview of different alternative black liquor recovery technologies, see for example Whitty and Verrill (2004).

In the search for alternative methods for energy and chemical recovery in the chemical pulp industry, more than 20 different gasification technologies have been studied over the years. In Sweden, for example, another black liquor gasification technology, besides the Chemrec technology, was developed by ABB during the 1990s. Technical problems, lack of funding and little interest from the market have resulted in only two technologies, the Chemrec and TRI processes, remaining for development effort during recent years. (Modig, 2005)

3.4.3 The early development of the technology

During the development of the Chemrec technology, several different plants have been operated during the last decades (in Hofors, Frövi, Skoghall, New Bern and Piteå). The development plant in Piteå was taken into operation in 2005. (Bergek, 2002; ETC, 2011)

The development has gone from non-pressurized air-blown gasification to pressurized oxygen-blown gasification. Since both firing in a gas turbine for electricity generation and motor fuel synthesis occur at high pressure, it is advantageous if the synthesis gas is already pressurized. Furthermore, pressurized gasification offers the advantages of smaller equipment and the ability to produce low- and medium-pressure steam from gas cooling, which can be used in the pulping process. (Stevens, 2001; Whitty and Nilsson, 2001; Landälv, 2007)

Air-blown gasification can be used if electricity generation is envisaged. However, the gas from air-blown gasification has a low heating value because it contains significant amounts of inert gas (mainly nitrogen), which requires modifications of the gas turbine combustion unit. Furthermore, it is very difficult to start the gas turbine system and maintain stable operation. These problems do not occur if oxygen is used instead of air for the gasification. In this case, the gas obtained has a higher heating value as it contains less inerts and no structural changes to the gas turbine system are needed, which makes the plant simpler and cheaper. Moreover, the equipment can be made smaller because of the reduced volume of gas. Motor fuel production requires oxygen-blown gasification in order to obtain a gas with a sufficiently high heating value for fuels synthesis. (Whitty and Nilsson, 2001; Marbe, 2005; Landälv, 2007)

The pressurized oxygen-blown version of the Chemrec technology, currently developed in Piteå, is primarily intended to replace the recovery boiler at a mill. Chemrec also has a booster technology, which allows for capacity additions in a mill where the recovery boiler is already running at maximum capacity, designed to handle the extra black liquor formed as a result of the increase in capacity. The booster technology is simpler; it is not pressurized, uses air as gasification agent, and produces a fuel gas that is used for steam production, which means that it can be considered as a commercial technology. As early as 1995, when the U.S. company Weyerhaeuser wanted to increase production at its mill in New Bern, USA and invested in a Chemrec booster, the facility was able to supply green liquor with an acceptable quality (the green liquor may be of lower quality because it only represented a small part of the total green liquor). The facility has had some technical problems over the years, including material problems in the gasifier, which led to closing of the plant in 2000. The plant was rebuilt and resumed operation in 2003. In 2008 the plant was again closed, this time due to decreased production at the mill, and has not resumed operation since. (Whitty and Nilsson, 2001; Chemrec, 2005a; Rudberg, 2006; Landälv, 2007; Furusjö, 2011)

3.4.4 Research and development at the plant in Piteå

The plans for a development plant in Piteå started in the second half of the 1990s. During the 1990s, black liquor gasification received political attention as a possible technology for increased electricity production in the pulp and paper industry which could contribute to increased renewable electricity production in Sweden. This finally led to a Swedish government body deciding to support construction of a BLGCC development plant. (Bergek, 2002; Tegnér, 2007) For a number of reasons, such as changed ownership structure in both Chemrec and the mill in Piteå, the plans were delayed and restricted to a plant for gasification and gas cooling. (Bergek, 2002; Landälv, 2007) Some of the reasons for which Piteå was selected as location for the development plant were significant interest from the mill owner (a kraftliner mill now owned by Smurfit Kappa) for the black liquor gasification technology and availability of a suitable space for the plant at the adjacent Energy Technology Research Centre ETC Piteå (Energitekniskt centrum i Piteå) (Landälv, 2007).

The plant, which started operation in September 2005, has a capacity of 3 MW black liquor (20 ton dry solids/day), corresponding to approximately 1% of the black liquor produced at the mill (Chemrec, 2005b; Rudberg, 2006). In the development plant in Piteå the standard gasifier operating conditions are 29 bar, 1000°C and the black liquor from the mill has a normal dry solid content of 73% (Landälv et al., 2010).

In 2004, a research program about black liquor gasification was started, the Black Liquor Gasification Program, which was a continuation of a program that started in 2001. During the first phase of the program (BLG I), 2004-2006, the task was to build, commission and test the development plant in Piteå, and perform fundamental research on a number of issues connected to the technology. During the second phase (BLG II), 2007-2010, the overall goal was to remove scientific obstacles to commercialization of black liquor gasification, to understand the process and to place it in its context within the pulp mill. Research has been conducted at several universities and research institutes in Sweden with financial support from agencies and foundations, as well as industry. (Landälv, 2007; ETC, 2011) This PhD project was partly funded by BLG II.

The program worked on, for example, challenges regarding materials for the gasifier, which has resulted in materials that can handle several years of operation. The problems with increased concentration of non-process elements are not as great as was expected, and there are different possible solutions to the problem. The results from the plant indicate an increase of the need for CaO, and correspondingly also the load of the lime kiln, by approximately 33%. (ETC, 2011) This is lower than predicted in, for example, (Ekbom et al., 2005), where an increase of the load of 41% was assumed. The goal is to limit the increase to 20-25% (ETC, 2011).

An important objective for the development plant in Piteå has been to demonstrate stable and continuous operation (Rudberg, 2006). Development of the recovery boiler technology has led to increased availability. In the U.S., Finland, Sweden and Norway, the availability of recovery boilers is above 99.5%, and mill owners expect similar availability for gasification units. One of the most important design criteria for the recovery boiler is thus high availability. Since pulp production is a continuous process, it sets high standards for each process unit to work with almost no errors. (Modig, 2005)

The development plant in Piteå has been in operation for 12,000 hours in total. The availability has gradually increased and during 2009 it reached approximately 70% on a monthly basis. However, the disruptions have been dominated by faults that would not have caused stops on a plant in full scale with better equipment. (ETC, 2011) In for example Ekbom et al. (2005), who assess the potential of possible future full-scale “N^{th}” BLGMF plants, it is assumed that the mill invests in four gasifiers and that three gasifiers are in operation at any given time, with the fourth initially of interest on standby.

As has been described, black liquor gasification was initially considered mainly as a technology for increasing power generation in the pulp and paper industry. In the beginning of the 2000s, however, Chemrec began to look at other possible usage of the syngas, including production of motor fuels, and during the last decade the focus has been shifted towards future implementation of BLGMF plants rather than BLGCC plants (Landälv, 2007). However, the BLGCC concept could naturally still be interesting; future energy prices and policy instruments will determine the concept that is most profitable. The focus in the development of black liquor gasification has been, as described, on the gasification and gas cooling steps. The processes for cleaning and processing of the gas and synthesis of fuels such as methanol and DME are based on known, commercial technologies. However, for example, small differences in gas composition can be an important factor, and it is therefore important to demonstrate the whole process from black liquor to motor fuel (Järås, 2006). The two main goals of BLG II, which were achieved during the program period, were to be able to generate syngas that could be cleaned with known technologies and used for synthesis of methanol and DME, and to generate input data for upscaling to industrial scale (ETC, 2011).

3.4.5 Current situation

In September 2010 a BLGMF demonstration plant was inaugurated in Piteå. The plan is to start operation during spring 2011. The plant converts the raw syngas from the gasifier, which was previously flared, into DME. The plant provides trucks that have been adjusted for operation on DME (designed by Volvo) with fuel for commercial test operation during approximately two years. The project has been partly financed by EU and the Swedish Energy Agency. (Landälv et al., 2010; ETC, 2011)

Chemrec is also planning for a full-scale BLGMF plant at Domsjö Fabriker in Örnsköldsvik, Sweden. The goal is to start production of DME for use in heavy trucks and methanol for blending in gasoline in 2014. The final investment decision is planned for autumn 2011. (ETC, 2011) The project has received a grant from the Swedish Energy Agency that amounts to 500 MSEK, which has been approved by the European Commission. The total budget of the project is approximately 3000 MSEK. (SEA, 2011) The Domsjö mill is a sulphite mill with two old boilers for energy and chemical recovery that are in need of replacement. The plant is to have 3 × 50% gasifier trains, each designed to gasify approximately 550 tds/day (corresponding to approximately 100 MW). (Landälv et al., 2010; Furusjö, 2011) Thus, it is possible that the first full-scale plant for the Chemrec technology will be based on liquor from a sulphite mill and not black liquor from a kraft pulp mill, which has been the main fuel feedstock used in development of the technology. However, as mentioned, these liquors are similar.

Based on the current situation for the development of black liquor gasification, it was assumed in this thesis work that large-scale implementation of full-scale black liquor gasification plants is unlikely to occur before around year 2020.

3.5 Availability of black liquor

The total production of black liquor in the world was approximately 670 TWh/year in 2005. A few countries dominate the production. In Europe, the total production is approximately 120 TWh, with 2/3 of the production almost equally distributed between Finland and Sweden. North America (USA and Canada) is the main producer with more than half of the world’s production. Asia is the third largest producer, with Japan as the largest producer (60 TWh). In South America, the largest producer is Brazil (30 TWh). (Ekbom et al., 2005)

The potential road transport fuel replacement share for motor fuel (methanol, although similar results could be expected for DME) production from black liquor gasification is greatest in Finland and Sweden, where approximately 1/2 and 1/4 respectively could be replaced. In USA, which has the largest black liquor production in the world, but a significantly lower production per capita than Finland and Sweden, the replacement share is not more than a few percent. (Ekbom et al., 2005) It should be noted that if motor fuels are produced via black liquor gasification, the mill will have a net usage of wood fuel and electricity compared to operation with a recovery boiler, as described in Section 3.2.

4 Factors influencing economic performance and CO_2 emission balances

The economic performance and CO_2 emission balances for possible future technologies such as black liquor gasification, other pulping biorefinery technologies or other biofuel technologies are dependent on a number of different factors. This chapter describes and discusses the factors studied in this thesis that were introduced in Section 1.1.

Some of the studied factors are more general, and are important when evaluating many different kinds of new technologies. Some of the factors are more specific for black liquor gasification. However, similar factors can be important for other technologies.

The general factors include for example how to evaluate the CO_2 emission balances of biomass conversion technologies that could be implemented in the future. There are a number of different methodological approaches for this (see Section 4.9). The possible development of the energy market and different policy instrument systems will influence the CO_2 emission balance estimations, as well as the economic performance (see Section 4.10). To make estimations of investment costs is very difficult, especially for future technologies that have not yet reached commercial status. For very large investments such as black liquor gasification, relatively moderate changes of the estimated investment cost could significantly influence the economic performance (see Section 4.8).

The number of possible final products that can be synthesized from gasified black liquor, or other gasified fuels, is large and includes for example electricity and different biofuels (see Section 4.1). Using different types of biofuels in motor vehicles requires different degrees of changes to the existing distribution infrastructure and existing internal combustion engine technologies. Black liquor gasification could be implemented in different types of mills (market pulp mills or integrated pulp and paper mills) with varying steam requirements (see Section 4.2). The applicability and performance of new types of technologies vary for different types of mills. The mill's need for investments in the near future is important to consider (see Section 4.3). For the case of black liquor gasification, the most relevant is whether or not the recovery boiler has reached the end of its technical lifetime and must be replaced. For the evaluation of black liquor gasification it is essential to compare with other alternative major investment options for kraft pulp and paper mills. These investments could be based on both conventional and new technologies (see Section 4.4). The commercial availability of CCS technology and its applicability for given pulp mill configurations could significantly influence both the CO_2 emission balances and economic performance of both mills with black liquor gasification and recovery boiler technologies (see Section 4.5). Mills will have either a surplus of steam that could be used in different ways, or a deficit of steam that has to be covered in some way (see Section 4.6). Increasing the degree of heat integration in order to decrease the mill steam demand could improve the profitability of black liquor gasification (see Section 4.7).

The influence of choice of key black liquor gasification process parameters, such as gasification pressure and temperature, choice of gas cleaning processes, gas turbines or fuel synthesis, and the performance of these sub-processes, are not included in this thesis work. Mass and energy balances for the process steps from black liquor to final product are essentially based on previous work. In order, for example, to examine the consequences of different gasification temperatures, detailed modelling/simulation of the entire BLGCC or BLGMF process is necessary, since the gas composition from the gasifier is influenced by the gasification temperature. This was beyond the scope of this thesis work. However, simulations have been conducted, primarily in Papers V and VI, in order for example to investigate the consequences of including CO_2 capture in a BLGCC plant.

4.1 Choice of product from gasified black liquor

In this thesis, black liquor gasification with production of either biofuels or electricity is considered. Production of bulk chemicals such as ammonia is also possible, but not included in this thesis.

In this thesis, electricity generation in a gas turbine combined cycle, as described in Section 3.3, is considered. Contrary to steam turbines, which can be tailor-made for each specific application, gas turbines are produced in certain standard sizes (Modig, 2005). A perfect match between the syngas flow and gas turbine capacity can be hard to achieve. However, in this work it is assumed that the gas turbine is sized to match the available syngas flow. The resulting differences due to this assumption, adopted in most studies, and the performance of a fixed-size gas turbine operating at off-design conditions have been quantified by Harvey and Facchini (2004).

There are, as mentioned, a number of possible biofuels that could be produced through gasification. Gasification is one of the two main routes for production of transportation fuels from a lignocellulosic feedstock, termed advanced or second-generation biofuels, together with lignocellulosic ethanol. Even if these technologies have not yet reached commercial status, the hopes are great that second-generation biofuels will reach high energy and cost efficiency and that they will be able to contribute substantially to the reduction of GHG emissions (see for example European Commission, 2006; IPCC, 2007). The biofuels available today, termed conventional or first-generation biofuels, include, for example, ethanol from sugar or starch crops and biodiesel from esterified vegetable oil. A number of life cycle analysis (LCA) and well-to-wheel (WTW) studies have been conducted for first-generation biofuels, and the results regarding possible GHG emissions reduction and energy efficiency are far from unanimous (see for example Delucchi, 2006; Larson, 2006) . Despite the wide range of results it can be concluded that the total potential for GHG emissions reduction from first-generation biofuels is low in the long term, due to high land area requirements and low cost efficiency (Hamelinck and Faaij, 2006; Larson, 2006).

When comparing biofuels there are many important aspects besides climate impact and economic performance that should be considered, such as other environmental impacts, the need for changes in infrastructure and total market penetration potential.

Distribution, storage and refuelling of liquid fuels, such as methanol and FTD, are easier and require less energy compared to gaseous fuels such as DME and hydrogen. DME can, however, be liquefied at normal temperature by pressurizing to about 5-10 bar. (Ahlvik and Brandberg, 2001; Ahlvik and Brandberg, 2002)

Methanol can be used both in modified gasoline and diesel engines. It can also be used as a low blend in gasoline and in fuel cells equipped with a pre-reformer. FTD can be used in conventional diesel engines and can be blended with conventional diesel. There is also ongoing technology development of specific FTD engines that can achieve higher efficiency. DME is the biofuel for diesel engines that is often ranked as the most promising from both an efficiency and emission perspective. DME cannot be used in conventional diesel engines; modifications are necessary. Similarly to methanol, DME can be considered for fuel cell vehicles. Hydrogen can be used as fuel in both modified gasoline and diesel engines, but the most important future use of hydrogen as fuel, and which achieves maximum efficiency, is in fuel cells. Fuel cell-based powertrains are believed to have a potential to achieve significantly higher efficiency than powertrains based on internal combustion engines. (Ahlvik and Brandberg, 2002)

Many factors will determine which biofuels will come to dominate in the future. The level of policy support needed in order to stimulate large volume production of biofuels will be influenced by the portfolio of fuels that are available for commercial production and usage. In this thesis DME, used in modified diesel engines, is chosen as an example of a possible future biofuel that can be produced world-wide on a large scale via gasification of solid biomass, and if available, from black liquor. DME is considered a fairly realistic option for investments around year 2020, unlike hydrogen where a whole new distribution infrastructure has to be established with large challenges associated with storage. Furthermore, in order for hydrogen to reach maximum efficiency and be superior to other fuels, fuel cell engines must become fully commercially available.

Previous studies generally indicate good profitability and a significant potential to reduce global CO_2 emissions if BLG with electricity or motor fuel production is implemented at kraft pulp mills. However, in the study by Modig (2005) co-occurrence of several different key parameters was required in order for a BLGCC plant to be profitable (see further Section 4.2). Larson et al. (2006) and Joelsson and Gustavsson (2008) showed that the potential to reduce CO_2 emissions, per unit of biomass used, is greater for electricity production than for production of motor fuels such as DME, methanol and FTD via black liquor gasification. However, Andersson (2007) showed that introduction of CCS in the power generation sector could change this. Larson et al. (2006) showed that whether biofuels or electricity production is the most profitable option is dependent on future energy prices. This study is the only previous study, of the studies presented in Chapter 2, which compares profitability of BLGCC and BLGMF plants. However, the study is done for North American conditions, which differs from the European conditions that are considered in this thesis (see Chapter 5).

In Paper I, DME via gasification of black liquor is one of the examples used to illustrate the system expansion method that is used for estimation of CO_2 emission balances in this thesis. Paper II compares the CO_2 emission balances for DME, methanol, FTD and electricity from BLG. In Papers III and IV, only DME is considered, while Papers V and VI consider both DME and electricity. Thus, Paper II, which does not include any economic analysis, is the only paper where more than one biofuel is considered. Comparison of different biofuels from an economic point of view is not included in this thesis.

No combinations of BLGCC and BLGMF plants are considered in this work. Thus, the BLG plant is assumed to be designed either for maximized electricity or motor fuel production. However, as described in Section 3.2, purge gas from the motor fuel synthesis can be used as fuel for electricity production in a gas turbine. This is considered in Paper V.

4.2 Mill steam requirements

In this thesis, both market pulp mills and integrated pulp and paper mills are considered. Few biorefinery concepts necessarily affect operation of the fibre line. Evaluation of the studied biorefinery cases is done by comparing the energy and material flows for the studied biorefinery case with a mill reference case (see Section 4.4). Thus, the pulp wood and pulp/paper streams are identical for the two concepts to be compared and whether the final product is pulp or paper is not relevant for the assessment of black liquor gasification, or other pulping biorefinery concepts. However, the steam demand is generally greater for integrated pulp and paper mills compared to market pulp mills, and will thus affect the applicability and performance of different biorefinery concepts.

In this thesis, different host mill types are considered, ranging from an integrated pulp and paper mill with a high steam use to a market pulp mill with a low steam use. Model mills representing both average Scandinavian mills and mills based on best available Scandinavian technology (technology in use at mills today) have been developed within the national Swedish research programs KAM (Eco-cyclic pulp mill) and FRAM (Future resource adapted pulp mill) (KAM, 2003; FRAM, 2005).

An average Scandinavian integrated fine paper mill has a steam use of 19 GJ/ADt (Delin et al., 2005a). Implementation of BLG on a large scale is unlikely to occur before around year 2020, as discussed in Section 3.4.5. Considering the increasing focus on energy efficiency, we expect that the current average steam use for a mill will be considered as high steam use a decade into the future. A market kraft pulp mill based on best available Scandinavian technology has a steam use of 11 GJ/ADt (KAM, 2003; Delin et al., 2005b). With state-of-the-art equipment and a higher degree of heat integration, it is possible to further decrease the steam use (Algehed, 2002; KAM, 2003). The levels of an average Scandinavian market kraft pulp mill (14.3 GJ/ADt) and an integrated fine paper mill based on best available Scandinavian technology (15.5 GJ/ADt) are in between these values (Delin et al., 2005a; Delin et al., 2005b). Thus, by considering mill steam demands ranging from below 11 GJ/ADt up to 19 GJ/ADt we cover a broad spectrum of mill energy usage, from a future market pulp mill with a low steam use to a future integrated pulp mill with a relatively high steam use. Since we expect current average steam use to be regarded as high, we expect current best steam use to be standard practice, or close to standard practice, a decade into the future. Thus, when discussing future market pulp mills and future integrated pulp and paper mills in general terms, we refer to mills with a steam demand of around 11 GJ/ADt and around 15.5 GJ/ADt respectively. The shift from a steam deficit to a steam surplus is somewhere around 14-15 GJ/ADt. Thus, mills with a steam use above this have to use additional fuel, besides black liquor, in order to cover the steam demand, and mills with a steam use below this have a steam surplus that can be utilised in different ways. The mill steam use levels discussed above are valid for mills using softwood as raw material.

Mills with different steam requirements have been considered in several previous studies that focus on detailed process modelling and energy analysis, for example in Berglin et al. (1999). However, few studies that include calculations of economic performance or CO_2 emission balances highlight the influence of mill steam requirements. Modig (2005) showed that the steam requirement of a mill significantly influences the economic performance of a BLGCC plant. For a mill steam demand of around 15 GJ/ADt, introduction of a BLGCC plant was not found to be profitable, whereas for a mill steam demand of around 10 GJ/ADt it could be profitable. The reason for this result is that the increase in total efficiency (power and heat production) between the mill with a BLGCC plant compared to the mill with a recovery boiler-based powerhouse technology is greater for a mill steam demand of 10 GJ/ADt compared to a mill steam demand of 15 GJ/ADt. This can be explained by the relatively low total efficiency for the recovery boiler-based powerhouse serving a mill with a steam demand of 10 GJ/ADt due to production of power in a condensing steam turbine unit. In Edwards et al. (2007) and Renew (2008), comparing different routes for production of biofuels, data concerning BLGMF plants integrated with a future market pulp mill (steam demand around 11 GJ/ADt) are used. That this is the case is however not clearly stated in any of these studies, and the results are presented in such a way that they appear to be general for all types of kraft pulp and paper mills.

In Papers I and IV a market pulp mill based on best available technology (BAT) is considered. Paper II considers both a market pulp mill and integrated pulp and paper mill based on BAT. In Papers III and V, a wide range of mill steam demand levels is considered. In Paper VI, a market pulp mill based on average Scandinavian technology is considered (several energy efficiency measures which considerably lower the mill steam demand are also considered).

The size of the mill is relevant when assessing the potential for black liquor gasification, since the specific investment cost for the technology decrease significantly with size. The tendency in the pulp and paper industry is fewer mills with larger production capacity (CEPI, 2008). This means that some mills will be closed down, while the remaining mills will increase their production capacity. In Papers I-V, the mill production capacity is 2000 ADt pulp/day. In Paper VI, the production capacity is 1000 ADt pulp/day. Both a constant and an increased production capacity are considered for the latter.

4.3 Mill investment requirements

In order for mills to consider implementation of full-scale BLG plants, the recovery boiler has to have reached the end of its technical lifetime. A recovery boiler has an average lifetime of around 30-40 years (Ekbom et al., 2003). In Papers I-V it is assumed that the recovery boiler is to be replaced and that the mill can choose between investing in a new recovery boiler or in a BLG plant. The option of investing in both a recovery boiler and a BLG plant is not considered in this thesis, mainly because the specific investment costs for both plants then would be significantly higher.

In Paper VI, it is assumed that the recovery boiler is not to be replaced. The mill could take advantage of a potential steam surplus (which could be created by implementing energy efficiency measures) and invest in a new plant for production of electricity or biofuels via black liquor gasification. It should be recalled that if part of the black liquor stream is processed in a BLGCC or BLGMF plant, less steam is generated per unit of black liquor compared to the recovery boiler. Thus, decreased steam demand in the mill provides the opportunity to invest in a BLGCC or a BLGMF plant without requiring purchase of additional boiler fuel to maintain the mill's steam balance. Both a constant and increased mill production capacity are considered. In the case of an increased production capacity, the mill can choose between rebuilding the recovery boiler to be able to process all the black liquor or, for example, investing in a BLG plant that can process the extra black liquor. The smaller BLG plants considered in Paper VI will obviously have a significantly higher specific investment cost compared to a full BLG plant.

Comparisons of different technologies for utilizing a potential steam surplus or debottlenecking the recovery boiler have been studied by, for example, Olsson et al. (2006) and Axelsson et al. (2006a). However, BLG with production of electricity or biofuels has not been considered as options for these purposes in previous studies.

4.4 Alternative investments for the mill

When evaluating black liquor gasification, or other pulping biorefinery concepts, it is necessary to define a reference case (alternative investment) to compare with. A natural choice of reference investment option is some form of "business as usual", where proven technology solutions are adopted. Thus, recovery boiler-based options where additional electricity is produced in the case of a steam surplus and a bark boiler is used in the case of a steam deficit (as described in Section 3.1).

Recovery boiler steam data has developed through the years, as discussed for solid biomass boilers in Section 3.4. Recovery boilers in average Scandinavian mills typically have steam data of around 60 bar, 450°C (Delin et al., 2005b). Until recently, the highest steam data in Scandinavians mills has been around 80 bar, 490°C, and this is the steam data considered in the model mills based on BAT developed within the KAM and FRAM programs (KAM, 2003; Delin et al., 2005b). In Papers I-IV, the steam data assumed for the reference recovery boiler investment option is 81 bar, 490°C. In Paper V, steam data that is likely to be standard for future recovery boilers is assumed: 112 bar, 540°C.

As mentioned in the introduction, in order to draw more general conclusions about the competitiveness of BLG, one must compare the technology with investment options based not only on conventional technologies, but also on other emerging technologies. However, it is natural if a study focuses on aspects of one technology, such as black liquor gasification, to relate only to a reference investment based on some sort of "business as usual". Then one has to be careful when drawing conclusions from the results. This is the case if, for example, the production cost of biofuels via black liquor gasification, compared to a mill reference investment option based on conventional technologies, is found to be lower compared to other production routes for biofuels.

Then one cannot automatically draw the conclusion that this is the best way to produce biofuels. From the mill's perspective, it could be other options that can generate a higher profit.

As mentioned in Section 3.1, there is a lot of ongoing research and development regarding extraction and usage of lignin. Extraction of lignin from black liquor has been tested at a pilot plant in Bäckhammar, Sweden (Berglin et al., 2010). The concept is based on addition of CO_2 to a black liquor side stream that is diverted from the evaporation plant, which results in lignin precipitation. The precipitated lignin is then filtered and washed. Implementation of lignin extraction influences the load of the recovery boiler. Thus, if investment in lignin extraction is made in connection with a substitution of the recovery boiler, the new recovery boiler could be smaller. If a mill wants to increase its production and the recovery boiler already operates at its maximum capacity, lignin extraction is a way to reduce the load of the recovery boiler. Extraction of lignin influences the load and design of the evaporation plant. For more details about lignin extraction, see Olsson (2009).

In previous studies, lignin extraction has often only been considered if the mill has a surplus of steam or if a mill is to increase its production capacity and the recovery boiler is a bottleneck (see for example Axelsson et al., 2006a; Olsson et al., 2006; Jönsson and Algehed, 2010). In such studies, lignin has been considered as a fuel and priced as wood fuel, and consequently it does not make sense to extract lignin if this means that wood fuel must be fired in order to compensate for the decreased steam production in the recovery boiler. However, lignin extraction could also occur with the purpose of selling/using the lignin as a feedstock for production of chemicals/materials instead of oil. The economic value of the extracted lignin is then related to the oil price. In these cases, it could make sense to extract as much lignin as is technically possible. Examples of chemicals/materials that could be produced from lignin are phenols, plastic materials and carbon fibres.

Eriksson (2001) considered lignin extraction, with the purpose of using lignin as a fuel, in connection with both recovery boiler- and black liquor gasification-based investment options. However, the study did not include any calculations of economic performances or CO_2 emission balances.

In Papers I-IV, the investment options for black liquor gasification are only related to reference investment options based on conventional technologies. In Paper V, black liquor gasification is also compared to extraction of lignin. Lignin is assumed to replace oil and as much as is technically possible is extracted. Furthermore, Paper V considers the possibility to implement CCS technology (see Section 4.5) as well as different systems for balancing a mill steam deficit (see Section 4.6). In Paper VI, black liquor gasification is considered as one option, together with for example extraction of lignin, for utilisation of mill excess steam. Lignin is here priced both as wood fuel and oil.

There is also ongoing research investigating extraction of hemicelluloses from the pulp wood, or from black liquor, to use as a feedstock for production of, for example, ethanol or different chemicals/materials such as acetic acid and polymers (see for example Brau, 2010). Work regarding extraction of hemicelluloses is still at the experimental stage.

4.5 Opportunities for CCS implementation at the mill

Black liquor gasification, and other gasification processes, offer possibilities to capture relatively large amounts of CO_2 at relatively low costs. CO_2 could be separated from boiler flue gases (including recovery boiler). The cost for the latter is, however, generally significantly higher. The captured CO_2 must thereafter be compressed and transported to a geological storage. Assuming that there is no CO_2 charge connected with the usage of biomass feedstock, including black liquor, the captured CO_2 must generate an income in order for CCS to be economically interesting. In this thesis it is assumed that captured green CO_2 generates an income corresponding to the charge for emitting fossil CO_2.

In a BLGMF plant, CO_2 is separated as part of the gas cleaning process, as described in Section 3.3. Therefore, only costs related to compression, transportation and storage of the CO_2 are incurred for a BLGMF plant with CCS compared to a plant without CCS. It should be noted that unless hydrogen is produced, almost half of the carbon is contained in the motor fuel product, and thus the amount of separated CO_2 is significantly lower than the amount that can be captured from the recovery boiler flue gases.

CO_2 is not separated as part of the gas cleaning process in the case of BLGCC. If CCS is to be considered, the gas cleaning could be changed to include a water-gas shift and removal of CO_2, as for a BLGMF plant. Thus, CO_2 is removed before the gas turbine, thereby reducing the gas flow in the gas turbine and downstream HRSG. The amount of CO_2 that can be captured in this case depends on the amount of gas that is shifted. Capturing of CO_2 from the gas turbine flue gases could also be considered. However, the concentration is low and the process requires large amounts of heat.

Capturing of CO_2 from the recovery boiler flue gases can be accomplished by absorption in different solvents such as mono-ethanolamine (MEA) or chilled ammonia (Hektor, 2008). Large amounts of heat are needed in order to regenerate the absorbent (desorption).

In order for CCS to be profitable for conventional coal power plants, which constitute very large emission point sources, the charge for emitting CO_2 has to be sufficiently high (Axelsson and Harvey, 2010). To compress, transport and store the CO_2 that is already separated as part of a gasification process can be profitable at a significantly lower level of the CO_2 charge. However, it can be discussed whether there will be a widespread infrastructure for storing the CO_2 if capture is not widely introduced in the power sector with its very large emission point sources.

Jönsson and Berntsson (2010) state that the majority of European kraft pulp mills are located far away from other large energy-intensive industries and potential fossil CO_2 capture clusters, and that further investigation is necessary to determine whether biomass-based CO_2 capture clusters, located in Scandinavia, are large enough to motivate investments in infrastructure for transportation of captured CO_2. Furthermore, the distances to possible storage location for these biomass-based clusters are generally long compared to the possible fossil CO_2 capture clusters in central Europe. Thus, even with a relatively widespread infrastructure for transportation and storage of CO_2 in central Europe, it is not at all certain that such infrastructure is located within the vicinity of most kraft pulp mills.

CCS in connection with black liquor gasification has been studied by, for example, Möllersten (2002) and Andersson (2007) who show that the potential for CO_2 emission reduction in connection with BLGCC and BLGMF systems can be significantly increased if CCS is considered. In all papers, CCS is considered as an option for the surrounding energy system. The opportunities for CCS at the mill are considered in Papers I, V and VI.

4.6 Choice of technology for balancing a steam deficit/surplus at the mill

A mill will have a net surplus or deficit of steam depending on the energy and chemical recovery technology selected, whether or not CCS and/or lignin extraction is considered, the degree of heat integration, etc. As described in Sections 3.1 and 3.2, the conventional way to utilize a steam surplus is to produce additional electricity in a condensing steam turbine unit, and the conventional way to cover a steam deficit is to use a bark boiler.

Other options for utilization of mill excess steam include integration with another industrial process with a net steam deficit, such as an ethanol plant (KAM, 2003). The steam could also be used for district heating delivery. However, excess heat at lower temperatures is a better choice for this purpose.

Integration with solid biomass gasification with production of electricity or motor fuels could be an option for mills with a steam deficit. There are currently a number of projects focused on development of solid biomass gasification technologies, as mentioned in Section 3.4.1. The main difference compared to black liquor gasification is that the feedstock must be more extensively pre-treated, for example dried prior to gasification. Different types of gasifiers are considered, including fluidised beds and entrained flow gasifiers.

Since there is a substantial steam/heat surplus from gasification processes, integration with other industrial processes or district heating systems can improve both the economic performance and the CO_2 emission balances of the process. Black liquor gasification is an integrated part of the pulp mill process, and the excess of steam/heat can be used all year round in the mill processes. For solid biomass gasification, however, there is no natural integration as in the case of black liquor gasification. There are a limited number of heat sinks that are large enough and that are able to accept excess steam/heat all year around. In countries like Sweden and Finland, the pulp and paper industry constitutes a significant integration potential for solid biomass gasification concepts. Several studies show that motor fuel production via gasification of solid biomass can be done more efficiently integrated with pulp and paper mills than in stand-alone mode (see for example McKeough and Kurkela, 2008; Joelsson et al., 2009). Furthermore, Wetterlund et al. (2011) showed that integration with a pulp and paper mill generally constitutes a more attractive option for solid biomass gasification plants compared to integration with a district heating system.

Integration of solid biomass gasification is primarily an option for integrated kraft pulp and paper mills, since kraft market pulp mills generally have a steam surplus, not a steam deficit. With increased energy efficiency, the integration potential for solid biomass gasification in pulp and paper mills decreases. As described in Section 4.2, an integrated fine paper mill based on BAT only has a relatively small steam deficit. However, implementation of new technologies such as black liquor gasification, lignin extraction or CCS increase the integration opportunity for solid biomass gasification, and it could also be an option for market pulp mills. One can nevertheless question whether it is realistic to consider investment options that require a mill to simultaneously implement several new technologies within a relatively short time frame. Thus, it might for example be more realistic to cover the steam deficit in a mill with a BLGMF plant with a conventional biomass CHP unit than with a solid biomass gasification plant producing motor fuels and/or electricity.

Larson et al. (2006) considered integration with solid biomass gasification for BLGMF plants integrated in integrated pulp and paper mills. However, it was shown that, generally, the profitability is higher if a bark boiler and steam turbine is used instead of a biomass gasification combined cycle. The option with gasification instead of combustion in order to cover a steam deficit was only considered if BLG was implemented and not for recovery boiler-based options.

In Papers I-IV, handling of a steam surplus/deficit is restricted to conventional technologies. Paper V also considers solid biomass gasification in order to cover a steam deficit. In Paper VI, the possible steam surplus is used for additional electricity production, CCS, lignin extraction or black liquor gasification. Since the opportunity for purchasing external wood fuel not is considered in Paper VI, CCS, lignin extraction and black liquor gasification are viewed as possible ways to utilize a potential steam surplus.

4.7 Degree of heat integration

There are a number of possible ways to save steam in a pulp mill. Common approaches include the following:

- Redesigning the heat exchanger network in order to remove pinch violations. By using pinch analysis (Linnhoff, 1993), the theoretical minimum heating and cooling demand of the pulp process can be determined. The pinch temperature divides the mill stream system into two parts, one part above the pinch where there is a heat deficit which has to be met by external heating (steam), and one part below the pinch where a heat surplus must be removed by external cooling. The net heat flow as a function of temperature can be represented by the grand composite curve (GCC). If, for example, heat is transferred from above the pinch to below the pinch in the existing heat exchanger network, both the heating and cooling demands become higher than the minimum target values (a pinch violation). By removing this pinch violation, both the heating demand (steam consumption), and cooling demand can be decreased.

- Switching to new energy-efficient equipment.

- Using excess process heat to replace boiler utility steam in modified process units. To be able to use the heat sources for this purpose, the excess heat temperature must be quite high and often a redesign of the secondary heat system is necessary.

In typical average kraft pulp and paper mills, there are large opportunities for steam savings using all of the above-mentioned options (Axelsson et al., 2006b). The BAT model mills developed within KAM and FRAM are designed with a heating and cooling demand close to the target levels established using pinch analysis (that is, there are practically no pinch violations). Furthermore, BAT implies energy-efficient equipment. However, there are of course other types of energy-efficient equipment that are commercially available for certain process units, but that were not yet in use at any Scandinavian mill at the time when the model mills were defined. The use of excess heat to replace live steam, for example in the evaporation or drying plant, was not considered in the construction of these mills either.

When new processes such as black liquor gasification are to be implemented at a mill, heat integration aspects are important. As was described in Section 3.2, BLGCC and

BLGMF plants have a net surplus of heat that can be used to produce process utility steam for the mill. Integration through the utility system for excess heat at required temperature levels could be preferable over direct heat-exchanging between streams for practical reasons, such as usage of only one heat exchanger in order to satisfy a heating or cooling demand and long distances between streams. Also integration within the BLG plant could partly be done through the utility system. Excess heat at temperature levels that are insufficient for production of process utility steam could, for example, be used in the mill's secondary heat system to preheat make-up boiler feed water and thereby decrease the amount of LP steam used in the feed water tank.

The opportunities for steam-saving measures at a mill are dependent on the energy efficiency of different process equipment, the design of the heat exchanger network, etc. The hot utility system of a pulp mill includes not only steam, but also warm and hot water at different temperature levels. Production of hot and warm water is handled in the secondary heat system together with make-up boiler feed water preheating. Process streams with a cooling demand are used as heat sources in the secondary heat system. The cooling demand that remains after heat recovery in the secondary heat system is usually handled by using cooling water, but could also be used as a heat source to produce district heating or to replace utility steam in modified process units. To be able to use the heat sources for these purposes, the temperature must be quite high. Traditionally, the secondary heat system is often designed with close to minimum heat transfer area and consequently most of the available high temperature heat sources are used (Wising et al., 2002). An alternative approach is to design the secondary heat system using heat sources at temperature levels that are just high enough for the water heating purposes considered. This releases more high-temperature heat that can be used for other purposes. This will however lead to increased heat transfer area due to smaller temperature differences. Whether the second design approach is better than the first is determined by the economic benefits of using the released high temperature heat sources in relation to the increased cost for heat transfer area.

Introduction of black liquor, or solid biomass, gasification could increase the net cooling demand and thus the available amount of mill excess heat. This could imply that the same amount of steam could be saved at a lower cost, that more steam could be saved for the same cost, or that it is possible both to deliver considerable amounts of district heating and at the same time use excess heat to replace process steam.

The consequences of, and therefore also the investment opportunity for, steam-saving measures are different for different mills. For mills with a steam surplus, lowering the mill process steam demand makes it possible to increase the electricity production. Thus, the associated investment opportunity is dependent on the electricity price. For mills with a steam deficit, lowering the mill steam demand could result in less wood fuel being fired in the bark boiler. Reduced steam production in the bark boiler results in reduced electricity generation (given that HP steam is produced and expanded in a steam turbine). Thus, the profitability of investment in steam savings is in these cases dependent both on the wood fuel price and the electricity price.

In all papers, the net steam surplus from the BLGMF or BLGCC plant is assumed to be used in the mill processes. In Papers I-III and V-VI, low-temperature excess heat from the BLG plant is used to preheat make-up boiler feed-water. In Paper IV, the effect of an increased degree of heat integration on the economic performance and CO_2 emission balances of DME production via BLG is investigated. A systematic comparison of different degrees of heat integration with respect to profitability and CO_2 emission balances has not been studied previously for BLGMF systems. The secondary heat system, including heat sources from both the BLGMF plant and the rest of the mill processes, is redesigned to be able to reduce steam usage in the evaporation plant. In Paper VI, where the calculations are based on an average Scandinavian market kraft pulp mill, different steam-saving measures are considered.

4.8 Level of investment costs

Estimation of investment costs is very difficult, especially for non-commercial technologies. In addition, it is often very difficult to understand exactly what is included in investment cost estimations in different published studies, and therefore a fair comparison of the different concepts is hard to achieve. For very large investments such as black liquor gasification, relatively moderate changes of the estimated investment cost could significantly influence the profitability.

Papers III and IV do not include any sensitivity analysis with respect to investment costs, whereas in Papers V and VI the investment costs for non-commercial technologies are increased by 30% and 25% respectively, to investigate the impact on profitability.

4.9 Methodology for evaluation of CO_2 emission balances

When evaluating the CO_2 emission balances or overall energy efficiency of introduction of new biomass-based technologies such as black liquor gasification, or other pulping biorefinery concepts, at pulp mills/pulp and paper mills, it is important to adopt a life cycle perspective and consider the impact of all steps from feedstock to final product(s). There are a number of different approaches that can be used for this purpose, and different choices can be made for each step from feedstock to product. Thus, different studies can come to very different conclusions about, for example, the climate effect for a given product and feedstock. These issues have been heavily debated, particularly regarding evaluation of different biofuel routes, as described in Section 4.1. The evaluation of energy efficiency and climate impact of biofuels and other transportation options is usually done from a well-to-wheel (WTW) perspective. A WTW study is a form of life cycle analysis (LCA) that is normally limited to the fuel cycle, from feedstock to tank, together with the vehicle operation, and that typically focuses on air emissions and energy efficiency (MacLean and Lave, 2003; Edwards et al., 2007). A WTW analysis generally does not consider the energy or the emissions involved in manufacturing energy conversion facilities, or end-of-life aspects. In this section, WTW analysis will be used to illustrate different methodological approaches and issues regarding the different steps from feedstock to product. However, the discussion can easily be generalised to apply to other products as well.

Figure 7 illustrates possible main energy and material flows between the main steps in a WTW analysis of biofuels, produced at a mill or other biofuel plant. If biofuel is produced at a mill, or the production is integrated with another industrial process, the flows represented are net differences compared to a reference investment option, as discussed in Section 4.2.

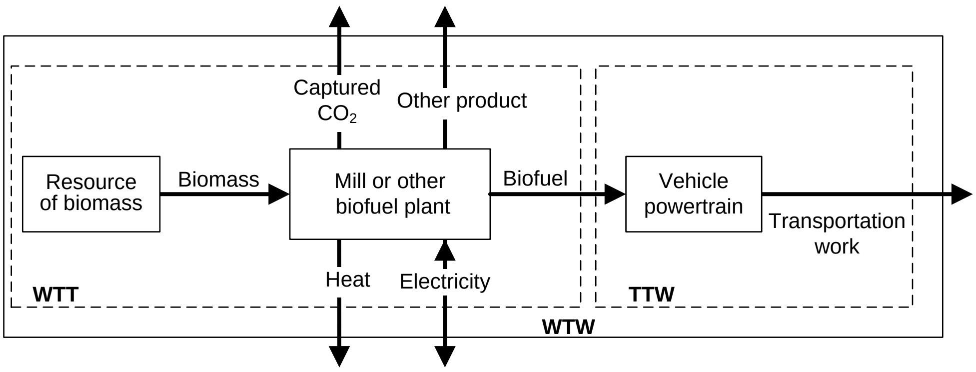

Figure 7. *Simple illustration of possible main energy and material flows between the main steps in a well-to-wheel (WTW) analysis of biofuels (from a mill or other biofuel plant) where also the well-to-tank (WTT) and tank-to-wheel (TTW) parts are illustrated.*

The first step in a WTW chain includes operations required to extract, capture or cultivate the primary energy source, in this case biomass feedstock. Thereafter, the biomass needs to be transported to the biofuel production plant. At the biofuel production plant, the biomass is processed into biofuel and possibly also other products such as electricity, heat or other by-products. The biofuel production plant may have a deficit of electricity. The biofuel production process may also have a net deficit of steam (an example is an ethanol plant as mentioned in Section 4.6). However, this is usually handled within the plant by firing external fuel, or by using internal by-products. Thus, the biofuel plant will not have a heat deficit. It could also be possible to capture CO_2 in the process, as discussed in Section 4.5. The produced biofuel is then distributed to refueling stations. The final step includes the vehicle operation where the biofuel is used to fuel the vehicle's powertrain. A well-to-tank (WTT) analysis includes the steps from feedstock to tank, and thus does not include the vehicle operation stage. This type of analysis could be used for example when comparing different ways to produce a specific biofuel. Most studies are focused on direct effects from physical flows in the

WTW chain, but some studies also include an estimation of contributions to system change (see for example Hillman, 2008).

4.9.1 By-product allocation

How to allocate the distribution of environmental burdens between the different outputs of a process producing more than one product has been one of the most controversial and heavily debated issues of LCA methodology, as it can have significant impact on the results (see for example Finnveden et al., 2009). Several reviews of WTW studies of various biofuels show that by-product allocation is one of the key issues that influence the GHG and energy efficiency results (Delucchi, 2006; Fleming et al., 2006; Larson, 2006; Börjesson, 2009).

Allocation can be done on the basis of physical properties (mass, energy content, volume, etc.) or on the basis of economic value. Allocation can also be avoided through system expansion or substitution, that is, expansion of the system's boundaries to include the additional functions of all by-products. By-product credits can sometimes also be handled by recalculating by-product streams into the same raw material as used for the main product and then subtracting the calculated amount from the raw material usage.

In Edwards et al. (2007) and Renew (2008), comparing the climate effect of different routes for production of biofuels, by-products are not handled consistently; system expansion and recalculation as well as physical allocation are used. Using physical allocation or recalculation to handle, for example, co-produced electricity results in the influence on the electricity production system not being reflected in the calculations.

4.9.2 Reference system

In system analyses of projects aiming at increasing production of biofuels, or other biomass-based products, with the purpose of assessing the potential to decrease global fossil GHG emissions or fossil energy use, a baseline or reference system must be defined, based on an estimation of what would have occurred in the project's absence. The reference system should include alternative pathways for the production of transportation fuel as well as for electricity, heat, and by-products (depending on whether allocation is considered). If the feedstock production results in land-use change, an alternative land use must also be included in the reference system. Similarly, when the same feedstock is in demand for other purposes an alternative biomass use should be included, as the increased use of a resource with constrained production volume results in less of that resource being available for other parts of the system, which can cause important indirect effects that may significantly affect the results (Ekvall, 1999; Ekvall and Weidema, 2004; Merrild et al., 2008).

The choice of reference system depends largely on the aim and time frame of the study. In general, the reference system should constitute a close alternative to the studied system, adopting the same technology level. Thus, if the study includes technology for which commercialisation is not imminent, the reference system should incorporate projected BAT for the same time frame rather than presenting average technology.

Another concern is the choice between average and marginal technologies for the reference system. A number of LCA-related publications recommend the use of a marginal approach for change-oriented studies of possible future systems, particularly for comparison between different systems (see for example Weidema et al., 1999; Tillman, 2000; Ekvall and Weidema, 2004).

Several studies, for example Hillman and Sandén (2008), show that the reference system selected results in a large degree of variation in the WTW CO_2 emissions, and that it may have consequences for the ranking order of the studied biofuels. This makes it reasonable to include several different reference systems (scenarios) in biofuel WTW studies, or studies of other biomass conversion systems, in particular when studies are made for a future situation (see also Finnveden et al., 2009). See further Section 4.10.

4.9.3 Functional unit

In studies where different systems are compared, the functional unit must be carefully selected and defined. When biofuels are compared to each other and/or to fossil-based motor fuels, the service provided – such as the distance travelled – can be chosen as the functional unit, as argued by for example Edwards et al. (2007).

If biofuels are to be compared with other bioenergy applications, another functional unit must be chosen. Several studies, for example Schlamadinger et al. (1997) and Gustavsson et al. (2007), emphasise the importance of considering the resource that will be limiting, for example for the GHG reduction potential. For bioenergy systems, this will typically be the available amount of biomass or the available land for biomass production. If the feedstock is the same in all considered cases, for example forest residues, the relative order of the results will of course be the same when reporting per ha and year as when reporting per unit biomass. When different feedstocks are compared, however, land use efficiency becomes increasingly important, since the land area available for biomass production is limited.

The choice of functional unit is associated with several methodological considerations. If, for example, the results are presented as driving distance per ha, adjustments of included processes need to be made by recalculation to the considered type of biomass. This may lead to the inclusion of unlikely components in the system studied.

If system expansion is used for a system with a relatively low biofuel output and a large output of a by-product, such as electricity, a high CO_2 emissions reduction potential may be erroneously attributed to the properties of the biofuel when it is really an effect of a large electricity output. To counter this problem, Schlamadinger et al. (1997) propose a method where the reference entity is expanded so that all studied systems produce the same output. A similar approach is used by Gustavsson and Karlsson (2006) who propose to expand the functional unit to include all energy carriers or products produced. Using the method of an expanded functional unit, however, may lead to the inclusion of unlikely components in the system studied. Further, when comparing very different systems or systems of a very complex nature there is a risk for losing transparency in the calculations.

4.9.4 General methodological approaches

As described previously, several methodological choices must be made when conducting a WTW analysis. When it comes to methodological issues such as choice of reference system and handling of by-products, one can discern a number of different approaches. A common approach in WTW studies is, for example, to use system expansion or substitution for crediting by-products, as recommended by the ISO standard (ISO, 2006). This is sometimes compared with other ways to handle by-products, such as allocation based on energy content or economic value. The reference system for electricity generation is often based on average, or sometimes marginal, technology, or a system using the same raw material as for the motor fuel production – and then the amount of raw material used for the electricity generation is added to/subtracted from the usage of raw material for production of the fuel.

Another approach is to use system expansion for all flows involved in the WTW chain. The flows entering or leaving the biomass conversion system are assumed to cause a change in the surrounding system. Since all flows are handled by system expansion, allocation of by-products need not be considered. This approach has been used by, for example, Andersson (2007) for comparison of different biofuels, as well as electricity, from gasified black liquor.

One approach is to expand the reference entity so that all studied systems produce the same output, as discussed in the previous section. For example, if a biofuel is produced together with electricity and district heating, it is compared to a reference system where the same amounts of transportation fuel/transportation work, electricity and district heating are produced. This method also avoids allocation of by-products. Joelsson and Gustavsson (2008) use this approach for comparison of different systems, including black liquor gasification-based systems, for production of biofuels and electricity.

To be able to present the results as driving distance per ha, all flows must be recalculated into the considered type of biomass. Thus, technologies in the reference system assumed for electricity generation, district heating, etc, must be biomass-based.

As is apparent from these descriptions, there are no sharp dividing lines between the different approaches. There are similarities, as well as clear differences, between the described methods. The first one is more a general description of conventional WTW studies, mixing elements from the other described approaches/methods. Whichever method is used, it is important that the reference system constitutes a close alternative to the studied system, using the same technology level.

Some of the approaches are only applicable to WTW studies of biofuels, or other biomass-based transportation options, whereas for example the system expansion method and the approach where all the studied system produce the same output are applicable for other biomass conversion systems, or other systems, as well.

4.9.5 Specific issues for the different energy and material flows

Unless fallow land or waste biomass is used, both direct and indirect land-use changes associated with biomass usage can cause large increases of CO_2 emissions. However, also for waste biomass, such as forest residues, soil carbon dynamics can have a substantial impact. When logging residues are removed from the forest, the soil carbon stock will in general be lower than if the residues were left in the forest to decompose, particularly if looked at over a short time period. The magnitude of the impact of the soil carbon decrease is, however, uncertain (Holmgren et al., 2007).

The supply of biomass is very much dependent on local conditions, and what kind is available will vary for different regions. Another important issue regarding the future supply is that not only the theoretical potential will matter; also technical, ecological and economic factors will have an impact. The fact that the supply of biomass is limited is acknowledged by several studies (Berndes et al., 2003; Hoogwijk, 2004; Lindfeldt et al., 2010). An increased use of a resource with constrained production volume results in less of that resource being available for other parts of the system; therefore efficient use is essential if the CO_2 benefit of substituting biomass for fossil fuels is to be maximised. Since biomass is a limited resource, it is not possible to solve the whole climate problem by substituting biomass for fossil fuels. To be able to give credits for biomass released for other use when implementing efficiency measures, for example, it is of crucial importance to take the marginal effects of limited resources into consideration when evaluating CO_2 emissions (see for example Axelsson, 2008).

If different pulp mill biorefineries are compared, such as electricity and biofuel production via gasified black liquor, biofuels can often achieve higher CO_2 emission reduction than electricity if limited availability of biomass is not taken into consideration; see for example Joelsson and Gustavsson (2008) and Larson et al. (2009). However, both of these studies emphasise the amount of biomass used, and that the potential to reduce CO_2 emissions, per unit of biomass, is higher for electricity generation. Andersson (2007) considers the limited availability of biomass by considering an alternative biomass usage.

How large emissions are and how much energy is needed for the transportation, handling and distribution, will be dependent on the type of biomass, the size of the production plant, and whether it is possible to supply the plant with biomass from the local region, or whether biomass must be transported from a larger area or even imported from another country.

A net deficit or surplus of electricity can be handled in different ways, as discussed in Section 4.9.1. In the case of electricity grids, one can use the average GHG or energy intensity of the entire system, the build margin or the operating margin (see for example Kartha et al., 2004; Schlamadinger et al., 2005; Ådahl and Harvey, 2007). What is a relevant grid electricity mix or marginal technology to use is dependent on, for example, the time frame and the system boundaries of the study. An electricity deficit or surplus can also be handled by assuming that the electricity is produced in a biomass-fired power plant. For production processes with a deficit of electricity, the calculated amount of biomass for electricity production is added to the amount of biomass feedstock, and vice versa for processes with a surplus of electricity.

In studies estimating CO_2 emission balances for biofuels or electricity production via BLG, average grid electricity mixes have been considered by, for example, Larson et al. (2003; 2006), whereas marginal electricity production technologies, mainly based on coal or natural gas, have been considered by, for example, Isaksson (2000), Möllersten (2002), Andersson (2007) and Joelsson and Gustavsson (2008). In Edwards et al. (2007) and Renew (2008), the net usage of electricity for the considered BLGMF process is recalculated into biomass and added to the net biomass usage. This approach is used by for example Ekbom et al. (2003; 2005), in order to calculate the biomass-to-biofuel efficiency. When doing this, the assumed biomass-to-electricity efficiency becomes important, as shown by Joelsson et al. (2009).

Biorefinery excess heat could be used for district heating production. However, in order for this to be possible the production plant has to be located within reasonable distance from a district heating system with a base load large enough to be able to accept all excess heat from the plant. The alternative district heating production is very much dependent on local conditions, such as the heat demand and availability of different fuels. For example, in a Swedish perspective a biomass CHP plant is often considered as a technique competing against industrial excess heat (see for example Jönsson et al., 2008). When excess heat replaces CHP heat, biomass is released for other uses. Thus, it is important to be able to attribute a CO_2 emission credit for the indirect contribution to a decreased use of biomass. In a European perspective, coal-based CHP could be considered as a technique competing against industrial excess heat (Axelsson and Harvey, 2010).

Even if the markets for other possible by-products, such as different chemicals, are not local – as is the case for heat – it is important to consider the size of the market. Hillman and Sandén (2008) point out that the size of by-product markets should be considered, and different by-product credits could be given depending on the degree of market penetration of the studied biofuel.

The possibility of CCS could affect the CO_2 emissions of a biofuel system, or other biomass conversion system, both directly – if CO_2 capture is possible in the production process and the plant is located near an infrastructure for CCS – and indirectly if, for example, CCS is implemented in coal power plants (lowering CO_2 emissions from grid electricity).

The final steps in the WTW chain include distribution, dispensing and usage of the biofuels. Today oil-based fuels, above all gasoline and diesel, totally dominate the transport sector and different biofuels are likely to replace these fuels. However, since crude oil is a considerably limited resource, the dominant transportation fuels of the future could be coal-based. For example, FTD produced via gasification of coal, with as well as without CCS, could be considered for the future reference transportation system. Most studies assume that produced biofuels replace gasoline and diesel, whereas for example Andersson (2007) and Edwards et al. (2007) also consider replacement of other fuels.

4.9.6 Methodology in this thesis

In this thesis, system expansion is used for all flows when evaluating the CO_2 emission balances of introduction of black liquor gasification and other pulping biorefinery concepts. The flows entering or leaving the biomass conversion system are assumed to cause a change in the surrounding system. A net surplus or deficit of electricity, for example, is assumed to affect the build marginal electricity production. It is assumed that if biomass is used at pulp mills/pulp and paper mills, less biomass will be available for other parts of the system. This is taken into account by assuming an alternative biomass usage. The methodology is further described in the next chapter.

The majority of kraft pulp mills are located in areas where the primary bioenergy source is the forest. Therefore, wood fuel, for example forest residues, is considered as feedstock. The impact of soil carbon decrease has not been taken into consideration. No GHG species other than CO_2 have been considered. Since the feedstock considered is forest residues, the results in most steps of the biomass conversion system will not be significantly affected by including other GHG species (see for example the results in

Edwards et al., 2007). But for grid electricity production based on coal, for example, the contribution from other GHG such as methane could be significant.

In Paper I, results of WTW CO_2 emissions using the system expansion method for different biomass-based transportation options are compared with the results from a conventional WTW study.

4.10 Future energy market conditions

The development of the energy market and different policy instrument systems will influence the reference system and thus also the CO_2 emission balances, as well as the economic performance, of introducing new technologies.

The influence of different assumptions regarding the reference system when estimating the CO_2 emission balances for BLG concepts have been considered by, for example, Isaksson (2000), Möllersten (2002), Andersson (2007) and Joelsson and Gustavsson (2008). It is mainly different assumptions regarding marginal electricity production technology that have been considered. However, Andersson varies the entire reference system, including also reference transportation technology and alternative biomass usage. The results presented by Andersson and by Joelsson and Gustavsson indicate a higher potential for CO_2 emission reductions for electricity compared to biofuels, with the exception of hydrogen, regardless of whether the marginal electricity production is coal- or natural gas-based (if limited availability of biomass is considered). However, Andersson shows that if CCS is introduced in the power sector, biofuels can achieve higher reduction.

In Paper I, the reference system and the emission baseline are varied systematically, thus covering a large number of possible future energy systems scenarios. Combinations that are considered as less probable are indicated. In order to maintain internal consistency in the scenarios used, a smaller number of energy market scenarios with interdependent parameters can be adopted, thus providing an effective way to systematically vary the reference system. Energy market scenarios have been used in previous studies (see for example Andersson, 2007; Axelsson, 2008; Hektor, 2008; Difs et al., 2010) in order to estimate both CO_2 emission balances and economic performance at different future energy market conditions.

The profitability of investments in BLG plants has been investigated by, for example, Ekbom et al. (2005), Larson et al. (2006) and Andersson (2007). In the extensive study conducted by Larson et al., which includes both investments in BLGCC and BLGMF plants, it is shown how the assumed energy price scenario and inclusion of policy instruments affect the ranking of different biofuels, as well as biofuels versus electricity.

In Papers II-VI, energy market scenarios including both energy prices and associated CO_2 emissions for marginal use of energy carriers are used. The methodology adopted, including energy market scenarios, is further described in the next chapter.

5 Methodology for evaluation of economic performance and CO_2 emission balances

This chapter describes the methodology used for evaluation of economic performance and CO_2 emission balances. Figure 8 shows a schematic representation of streams entering or leaving the type of biomass conversion systems that are studied in this thesis and how they interact with the surrounding system. In Papers II-VI the only biomass conversion process studied is the kraft pulp mill, whereas in Paper I lignocellulosic ethanol production and production of biofuels and electricity via solid biomass gasification are also studied. As discussed in Chapter 4, for the pulp mill biorefinery concepts studied, the flows considered are net flows compared to a reference investment option, and thus the flows of pulp wood and pulp/paper are cancelled out.

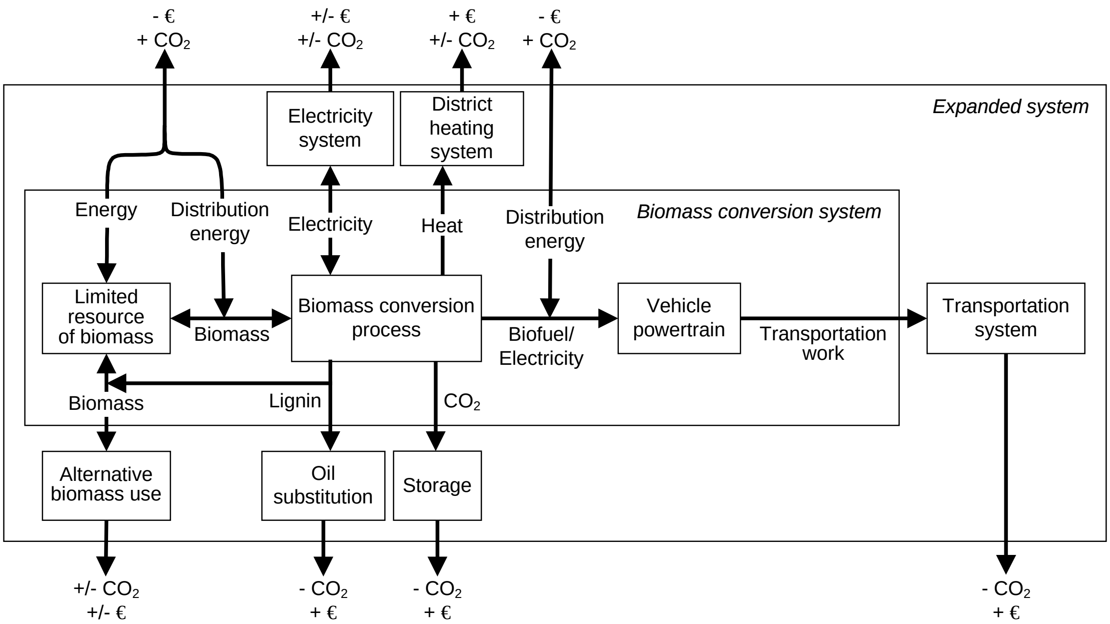

Figure 8. *A schematic representation of streams entering or leaving biomass conversion systems that are studied in this thesis and how they interact with the surrounding system. The CO_2 effect of each flow is indicated with +/-, where + means an increase and - means a decrease in CO_2 emissions. The economic value is also indicated with +/-, where + means a revenue and - means a cost.*

In this thesis a European energy system perspective is taken. Papers II-VI use energy market scenarios which have been updated in several steps during this PhD project. In this chapter the latest version of the scenarios is presented (see below). This version is used in Paper V and in the updated version of Paper IV. A similar version is also used in

Papers III and VI, whereas Paper II uses an older version of the scenarios. The CO_2 emission values used in Paper I are presented in the summary of the paper in Section 6.1.2.

It is assumed that there is a charge for emitting fossil CO_2. Whether the form of charge is a tax, purchase of tradable emissions permit, or similar is not vital for the calculations. The important assumption is that the CO_2 emission charge is assumed to be the same for all types of emitter.

In this thesis the biomass system is expanded to include alternative biomass use, by assuming that biomass used at the mill, or in any of the other biomass conversion processes studied, reduces the amount of biomass available for other applications in the system, thus increasing the CO_2 emissions from those applications. It is assumed that the high-volume user with the highest willingness to pay (WTP) will be price setting and thus constitute the alternative use. One potential marginal biomass user, considered in this thesis, is coal power plants where biomass can be co-combusted with coal. Other biomass users, such as boiler fuel substitution (oil) and industrial CHP, often have higher WTP for biomass compared to coal power plants. However, due to these users' limited demand, they are assumed not to constitute marginal high-volume users. If the mill has a surplus of biomass, the opposite effect is assumed and the amount of biomass available for other applications increases. Emissions and costs for collection, shipping and transportation of biomass are also considered.

In Paper I, coal power plants are considered as the alternative biomass user. The alternative where biomass is not subject to competition, and thus the use can be viewed as CO_2-neutral, is also considered in order to illustrate the marginal effect of biomass usage. In order to reach the goal for the share of renewable energy use within the transport sector setup within the EU, a dramatic increase in production of biofuels is needed. Hence, producers of biofuels could become a high-volume user of biomass. In the energy market scenarios, used in Papers II-VI, production of DME via solid biomass gasification (used as an example of a potential biofuel process) is considered, together with coal power plants as potential alternative biomass users.

A net surplus or deficit of electricity is assumed to affect the marginal electricity production. Since the timeframe for the biomass conversion projects studied is relatively long, base load build margin rather than operating margin is considered. The base load build margin is here defined as the type of electricity generation grid capacity addition affected by implementation of the biomass conversion project in question.

In Paper I, three state-of-the-art fossil electricity production technologies are considered as build margin technologies: coal power, coal power with CCS, and NGCC. In addition to these, fossil-free CO_2-neutral electricity production (for example, but not limited to, wind power) is also included as a possible future build margin technology. In the scenarios used in Papers II-VI, the main assumption concerning the electricity market is that the base load build margin production in the modelled time period will still occur in condensing plants fired with fossil fuel (Axelsson and Harvey, 2010). Coal power and NGCC plants, with as well as without CCS, are considered. The electricity price is assumed to be equal to the total cost of electricity generation for a new base load plant. The technology with the lowest production cost constitutes the base load build margin in each scenario.

There is assumed to be a policy instrument incentive scheme promoting production of green electricity (that is, electricity generation from renewable energy sources). Therefore, it is assumed that all produced electricity is sold and generates additional revenue from this policy instrument, whereas consumed electricity is purchased for the price of non-green electricity.

Oil-based fuels, above all gasoline and diesel, totally dominate the transport sector and different biofuels are likely to replace these fuels. However, as discussed in Section 4.9.5, since crude oil is a limited resource the dominating transportation fuels of the future could be coal-based. In Paper I, three different reference transportation technologies are considered: oil-based gasoline and diesel, and coal-based diesel with as well as without CCS in the production step (FTD via gasification of coal). In Paper I, vehicle efficiencies are included to be able to calculate the CO_2 effect of replacing a certain amount of transportation work. In Papers II-VI, only substitution of oil-based fuels is considered. In Papers III-VI, DME is assumed to replace diesel. The DME price (at plant gate) is set so that the end user will have the same cost per unit of fuel energy for DME as for diesel (it is further assumed that the vehicle efficiency is the same for DME as for diesel, and consequently the cost per km will be the same). Energy taxes and VAT are assumed to be the same for DME as for diesel, but diesel is subject to the CO_2 emission charge. However, DME is more expensive to distribute than diesel. As for electricity, there is assumed to be a policy instrument incentive scheme promoting production of biofuels.

Export of heat to a district heating system is only considered in Papers I and VI. It is assumed that if industrial excess heat is used, this affects the building of base load plants in the district heating system. The market for district heating is for natural reasons of a much more local character than, for example, the markets for biomass and electricity. Thus, it is not possible to define a single reference technology for district heating production in Europe under certain conditions, unlike for electricity generation. In Paper I, a biomass CHP plant is considered as alternative technology to industrial excess heat, whereas a combination of coal CHP and gas boiler is considered in Paper VI. The heat production cost of the alternative technology is assumed to determine the WTP for industrial excess heat.

Extraction of lignin is considered in Papers V and VI. In Paper V it is assumed that lignin replaces oil as a feedstock for production of materials or chemicals. Although not used for energy purposes, the lignin price is in this case set equal to the price for fuel oil (including the CO_2 charge) and is assumed to decrease CO_2 emissions as it would have done if replacing oil as a fuel. No additional policy instrument promoting production of green chemicals or materials is considered. Paper VI also considers lignin as a fuel, priced as wood fuel. In this case, exported lignin is assumed to increase the amount of wood fuel available for other applications in the system, thereby decreasing the CO_2 emissions from those applications.

As discussed in Section 4.5, captured and stored CO_2 from the mill is assumed to generate an income corresponding to the CO_2 emission charge. In Paper VI, the mill uses fuel oil in the lime kiln. This is not indicated in Figure 8.

5.1 Energy market scenarios

Table 1 presents the energy market scenarios used in Paper V and in the updated version of Paper IV. The scenarios used are constructed by using a tool developed by Axelsson and Harvey (2010). They reflect different future energy market conditions, and are based on two fossil fuel price levels (low and high) and two CO_2 emission charge levels (low and high) combined into four different scenarios. By using scenarios that reflect the strong connection between different energy market parameters, a packaged sensitivity analysis can be conducted. As stated, implementation of, for example, BLG on a large scale is unlikely to occur before around year 2020. It was therefore judged appropriate to use energy market conditions for 2030, representing an average year upon which to base estimations of cash flows and CO_2 emissions related to the different investment options.

Table 1. *Key data for the four energy market scenarios used for 2030.*

Scenario input		**1**	**2**	**3**	**4**
Fossil fuel price level[1]		Low	Low	High	High
CO_2 emission charge		Low	High	Low	High
	[€/t CO_2]	35	109	35	109
Green electricity policy instrument	[€/MWh]	26	26	26	26
Resulting values of prices and policy instruments [€/MWh]					
Electricity		68	90	74	98
Bark/by-products/wood chips[2]		27	52	30	56
Lignin		45	67	67	89
DME		56	77	88	108
Biofuel policy instrument		34	38	9	13
Resulting values of CO_2 emissions [kg CO_2/MWh]					
Electricity		679	129	679	129
(marginal production of electricity)		(CP)	(CP CCS)	(CP)	(CP CCS)
Biomass		262	301	262	301
(marginal user of biomass)		(CP/DME)	(CP/DME CCS)	(CP/DME)	(CP/DME CCS)
Lignin		295	295	295	295
(alternative feedstock)		(Oil)	(Oil)	(Oil)	(Oil)
Transportation		277	277	277	277
(alternative transportation fuel)		(Diesel)	(Diesel)	(Diesel)	(Diesel)

[1] Oil prices: Low: 74 USD/barrel, High: 126 USD/barrel.
[2] In the past years the prices of wood by-products and chips have been very similar.

As can be seen in Table 1, coal power (CP) plants constitute the marginal electricity production technology in all scenarios. In the scenarios with a low CO_2 charge, 1 and 3, coal power without CCS has the lowest production cost, while in scenarios with a high CO_2 charge, 2 and 4, coal power with CCS has the lowest production cost and thus constitutes the build margin technology. In Table 1 one can see how the electricity price, which is dependent on both the fossil fuel price level (coal price) and the CO_2 charge, varies between the different scenarios.

The DME price (at mill gate) is also dependent on both the fossil fuel price level (oil price) and the CO_2 charge. A higher oil price leads to a higher price for gasoline and diesel, which means that the gate price for DME increases. A higher CO_2 charge has the corresponding effect.

The coal power plant's WTP for wood fuel is dependent on the coal price, the CO_2 charge and the policy incentive value for green electricity, whereas the DME plant's WTP is dependent on the DME price (oil price and CO_2 charge), the policy incentive value for green electricity (the plant co-produces electricity: see Paper V) and the plant investment cost. In the scenarios based on a high CO_2 charge, the DME production plant sends separated CO_2 to storage and thus the WTP is also dependent upon the level of the CO_2 charge in these scenarios. In all of the scenarios, coal power plants have higher WTP than DME production. In order for Europe to have large-scale production of biofuels, the producers have to be able to compete for the biomass feedstock. Here, a policy instrument promoting production of biofuels is set at a level such that a stand-alone DME production plant will have the same WTP for wood fuel as a coal power plant. As can be seen, the required support is rather high in Scenarios 1 and 2, with a low fossil fuel price level, and rather low in Scenarios 3 and 4 with a high fossil fuel price level. The level of support for green electricity is set to represent an average value for Europe and is not varied between the scenarios. Another option would be to relate the level of the green electricity support to the level of CO_2 charge, assuming that a high CO_2 charge would give a lower support for green electricity since the two policy instruments, in principle, should benefit the same technologies in the electrical power sector.

The price of wood fuel in Table 1 is the selling price. When purchasing wood fuel, an additional cost for transportation is added (6 €/MWh). The variation of the transportation cost with the amount of purchased bark/wood fuel is not considered. The CO_2 effect of using biomass is calculated as an average value of the CO_2 effect of using biomass in a coal power plant and the CO_2 effect of using biomass for production of DME. In Scenarios 1 and 3, the CO_2 emissions reduction consequences of using biomass in a coal power plant are significantly higher than for DME production. However, in Scenarios 2 and 4, the CO_2 emissions reduction consequences of using biomass for DME production, now including CCS, are close to the CO_2 emissions reduction consequences of using biomass in a coal power plant. The CO_2 emission values for marginal users of biomass, presented in Table 1, are valid if biomass is exported from the mill. When importing biomass, CO_2 emissions for biomass conditioning (7 kg CO_2/MWh biomass) are added to the value in Table 1.

Paper V includes a sensitivity analysis where the policy instrument for green electricity is removed; thereby also influencing the price of wood fuel and the required policy instrument for DME (see Section 6.3.4). Papers III and VI use essentially the same energy market scenarios as presented here. However, the data concerning the DME plant is different and, for example, CCS is not considered. This strongly affects the required level of policy instruments for DME in Scenarios 2 and 4. Paper VI also includes a sensitivity analysis where the policy incentive values for electricity and biofuels are changed. In Paper VI, prices and CO_2 emission values for district heating are also included, and six scenarios for both 2020 and 2030 are considered.

5.2 General economic assumptions

The net annual profit (NAP) for each studied mill case is calculated by comparing revenues and costs. The annuity method is used to incorporate the investment cost in NAP. All economic calculations are performed using 2008 money value. Investment costs are adjusted using Chemical Engineering's Plant Cost Index (CEPCI). It is assumed that equipment costs are a function of scale given by $C/C_0=(S/S_0)^R$ where C is the investment cost of the equipment with size S, C_0 is the installed investment cost of the base size S_0, and R is the scale factor which determines how fast the cost per unit increase with size. For most units, R is 0.6-0.7.

The capital recovery factor (annuity factor) is set at values that represent a view of the investments as strategic. In Papers IV and V, it is set to 0.125 1/year, which for example is equivalent to an economic lifetime of 15 years and an interest rate of 9%. In Paper VI, the capital recovery factor is set to 0.2 1/year (including a sensitivity analysis where it is changed to 0.1). In Paper III it is set to 0.1 1/year. The cost for transportation and storage of CO_2 is set to 8 €/t (Hektor, 2008). The operating time is set to 7838 h/year (355 d/year with a mill availability of 92%).

6 Summary of papers

This chapter presents a summary of the papers included in this thesis. However, as described in the introduction, most aspects investigated in Papers II and III are also included in Paper V, and thus results from Papers II and III are not explicitly presented.

In Paper I, data for the different production routes was taken directly from literature, since the methodology is the main focus and not the comparison between the different cases. In the rest of the papers, however, much effort has been devoted to making the different concepts as comparable as possible, with the same assumptions regarding the key process units that are included in the different concepts. Data concerning mass and energy balances for the BLG and other pulping biorefinery concepts has been taken directly from or calculated based on data from previous studies. Further calculations were then conducted to establish energy and mass balances for these processes integrated with the studied mills. The possibility of polysulphide cooking (see Section 3.2) in connection with BLG was not considered.

6.1 Paper I

This section summarizes Paper I, "Implications of system expansion for the assessment of well-to-wheel CO_2 emissions from biomass-based transportation".

6.1.1 Aim and procedure

The aim of this paper was to show the impact of expanding the system to include the systems surrounding a biomass conversion system, when evaluating WTW CO_2 emissions for different biomass-based transportation alternatives. Four different cases are considered: DME via black liquor gasification (BLG:DME), methanol via gasification of solid biomass (BMG:MeOH), lignocellulosic ethanol (EtOH), and electricity from a biomass integrated gasification combined cycle (BIGCC) used in a battery-powered electric vehicle (BPEV). All four cases are considered with and without CCS. System expansion is used consistently for all flows. The results are compared with results from a conventional well-to-wheel study that only uses system expansion for certain by-product flows.

6.1.2 Input data

Table 2 presents input data for the different technology cases considered. All biofuels are assumed to be used in hybrid vehicles. Data for the BLG:DME case is based on Ekbom et al. (2003) and refer to integration with a future market pulp mill.

Table 2. *Input data for the plant configurations studied. Negative values indicate import to plant.*

		BLG:DME		BMG:MeOH		EtOH		BIGCC	
		no CCS	**CCS**	**no CCS**	**CCS**	**no CCS**	**CCS**	**no CCS**	**CCS**
Biomass feedstock	MW	-157[a]	-157[a]	-229	-229	-222	-222	-140	-140
Biofuel/electricity for use in transp.	MW	275	275	123	123	58	58	60	52
Electricity	MW	-101[a]	-111[a]	-13	-17	46	45	–	–
District heating	MW	–	–	13	13	88	88	60	61
Captured CO_2	kg/h	0	87,500	0	38,600	0	7,300	0	42,300
Biomass conditioning emissions	kg CO_2/ $MWh_{biomass}$	7.13		7.13		7.13		7.13	
Biofuel distribution emissions	kg CO_2/ $MWh_{biofuel}$	3.74		3.96		3.35		–[b]	
Vehicle energy use	MWh/ 1000 km	0.39		0.41		0.45		0.17	

[a] The incremental biomass and electricity use compared to the pulp mill reference case, that is for biomass the 125 MW forest residues imported to the plant plus the 32 MW bark that could be exported in the reference case, and for electricity the 56 MW (or 66 MW when CCS is considered) imported electricity plus the 45 MW electricity that could be exported in the reference case.
[b] Distribution efficiency is assumed to be 93%.

To highlight the influence of the surrounding system on the results, the reference system and the emission baseline are varied systematically, thus covering a large number of possible future energy systems. Table 3 shows the reference system matrix with CO_2 emission values for the different reference technologies considered.

Table 3. *Reference system matrix with CO_2 emission values.*

Biomass use[a] kg CO_2/$MWh_{biomass}$		Electricity[a] kg CO_2/MWh_{el}		District heating[a] kg CO_2/MWh_{heat}		Transportation[b] kg CO_2/1000 km	
Co-firing with coal (avoided emissions)	329	Coal	723	Biomass CHP	142	Oil	134
						Coal	289
						Coal w. CCS	160
		Coal with CCS	136	Biomass CHP	359	Oil	134
						Coal	289
						Coal w. CCS	160
		NGCC	374	Biomass CHP	271	Oil	134
						Coal	289
						Coal w. CCS	160
		CO_2-neutral	0	Biomass CHP	410	Oil	134
						Coal	289
						Coal w. CCS	160
No alternative use	0	Coal	723	Biomass CHP	-268	Oil	134
						Coal	289
						Coal w. CCS	160
		Coal with CCS	136	Biomass CHP	-50	Oil	134
						Coal	289
						Coal w. CCS	160
		NGCC	374	Biomass CHP	-138	Oil	134
						Coal	289
						Coal w. CCS	160
		CO_2-neutral	0	Biomass CHP	0	Oil	134
						Coal	289
						Coal w. CCS	160

[a] Well-to-gate values.
[b] Oil represents an average value between diesel and gasoline. Coal means FTD produced via gasification of coal. All fuels used in hybrid vehicles. Well-to-wheel values.

When heat from the biomass conversion processes substitutes CHP heat in a district heating system, biomass is released for other uses, in this case the alternative biomass usage described above. Note also that delivery of excess heat from the biomass conversion processes to a district heating network decreases the system's electricity generation potential, since the opportunity for power generation in the district heating network's CHP plant is decreased. Since hybrid vehicles are assumed for all analysed biofuels, the reference transportation technology is assumed to be hybrid fossil-fuelled vehicles.

6.1.3 Comparison study

The results are compared with the results from another WTW study, the JRC/EUCAR/CONCAWE European well-to-wheel study by Edwards et al. (2007) (hereafter referred to as the EU study), to investigate the effects of expanding the system. The results from the EU study are here considered in reference to all three marginal transportation alternatives included in this study. The EU study takes CCS into account for fossil-based systems but not for biomass-based systems. For this reason only the results for the studied technologies without CCS are compared with the EU study results. The EU study does not consider export of excess heat for district heating. A major methodological difference between the EU study and this study is the handling of electricity. In this study, electricity is managed by system expansion, which means that a non-neutral electricity balance affects marginal electricity production. In general, the EU study uses a substitution method for by-product allocation, which corresponds to system expansion. Electricity, however, is not considered a by-product[1]. Instead, for biofuel production processes with a non-neutral electricity balance, it is assumed that the electricity is produced in a biomass-fired power plant. Different electricity production processes with different efficiencies are considered for different biofuel routes[2].

6.1.4 Results and discussion

Figures 9 and 10 show the results. Since the different systems use different amounts of biomass, the results are presented per unit of biomass fed to the system. The striped bars indicate combinations that are considered to be less probable. For example, if CCS is implemented in the biofuel and BIGCC plants, it will probably also be implemented for transportation fuel production from coal feedstock. Similarly it can be assumed that if CCS is implemented in coal power plants it will probably also be implemented in plants producing transportation fuels from coal, where CO_2 is separated as part of the process. An electricity system with a CO_2-neutral build margin will probably be an indication of strong policy instruments promoting reduction of GHG emissions to the atmosphere. Hence, if the marginal electricity production is CO_2-neutral, a marginal transportation

[1] The reason for this is reportedly that the large credit that could occur if the electricity were deemed to replace marginal or average electricity would distort the results. This reasoning only occurs with regard to electricity.

[2] For the gasification of solid biomass, gasification-based electricity production with an efficiency of 42.5% is assumed. For cellulosic ethanol, a wood-fired steam turbine condensing power station with an efficiency of 32% is assumed. The same type of electricity production is considered for black liquor gasification, but with an efficiency of 40%.

technology based on coal (without CCS) is considered less probable. Implementation of CCS for coal-based electricity and/or transportation fuel production, in combination with biofuel production without CCS, could also be regarded as less probable. It has however not been defined as such here, since CCS is not yet as established for biomass systems as for fossil systems. Furthermore, as discussed in Section 4.5, the location of the plants might not be within the vicinity of an infrastructure for CCS.

Figure 9 shows the net CO_2 emissions compared to reference systems where the alternative biomass use is assumed to be co-firing with coal in power plants. The figure shows the CO_2 emissions for each of the four different marginal electricity production technologies considered, over a range representing the three marginal transportation technologies (oil represents the top of each bar). When the reference electricity production and transportation technologies are varied, the potential for CO_2 emissions reduction fluctuates, with several cases (particularly EtOH and BMG) showing little or no potential for CO_2 reduction.

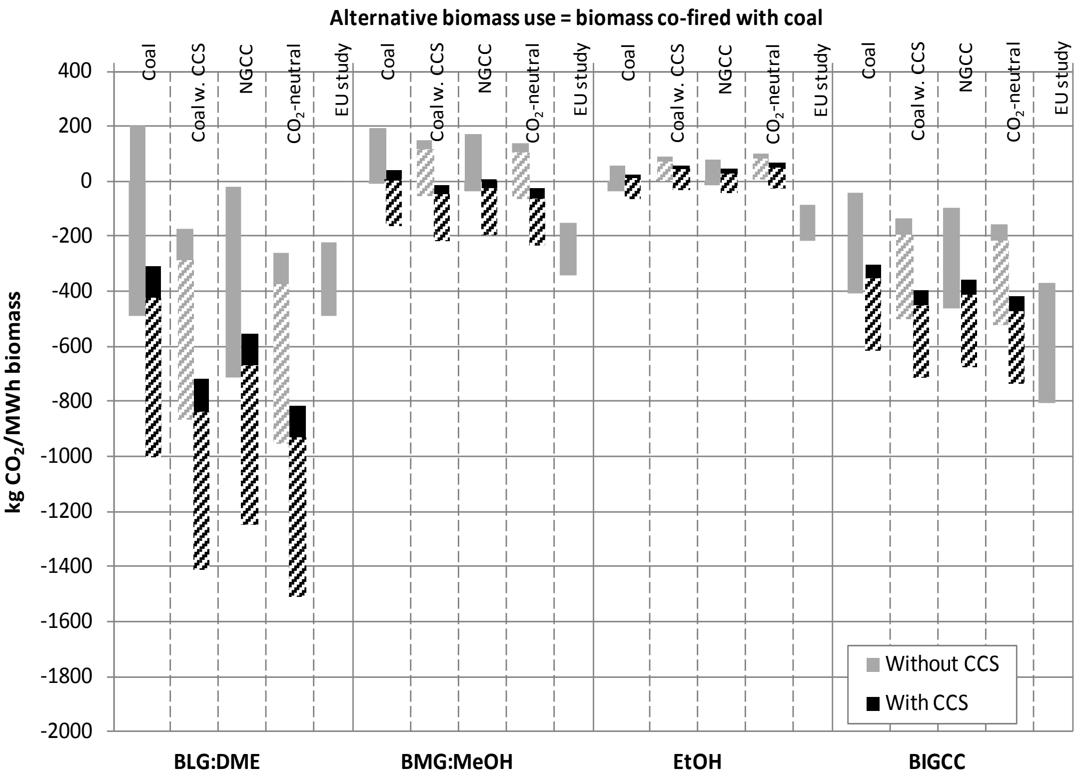

Figure 9. *Net CO_2 emissions per unit of biomass feedstock for the studied cases when the alternative biomass use is assumed to be co-firing with coal, for four different reference electricity technologies over a range representing the three marginal transportation technologies. The striped bars indicate combinations that are considered less probable. The results from the EU study, recalculated into CO_2 emissions per unit of biomass for the three marginal transportation technologies, are included for comparison.*

The influence of the electricity production technology differs between the technology systems studied, since they have very different energy balances (see Table 2). BLG, with a substantial electricity deficit, shows the largest variation and benefits from a low-CO_2-emitting electricity production technology (coal with CCS or CO_2-neutral electricity). EtOH, with a relatively high surplus of electricity, benefits from a high-CO_2-emitting electricity production technology (coal). However, as the large district heating delivery from the EtOH plant replaces biomass CHP heat which decreases the CHP electricity production, the electricity surplus from the EtOH plant is effectively almost cancelled out. The BMG process has a fairly low dependence on the electricity production technology, and thus shows similar results for all four electricity production technologies. For BIGCC, it was assumed that all the electricity produced is used in the transport sector. However, similarly to the EtOH process, the BIGCC delivers a large amount of district heating, which affects the alternative biomass CHP electricity production. In the same way as the BLG process, the BIGCC benefits from low-CO_2-emitting marginal electricity production.

The figure also includes the comparison results from the EU study for the cases without CCS. The EU study results have been recalculated from CO_2 emissions per vehicle km, to net CO_2 emissions per MWh of biomass for each of the three reference transportation technologies. As can be seen, the EU study in general shows a significantly higher potential for CO_2 reduction, with the exception of the BLG:DME system.

As expected, the cases with CCS show a considerably larger potential for CO_2 reduction than the cases without it – in particular for BLG, BMG and BIGCC, where the sequestrable amount of CO_2 is high.

Figure 10 shows the net CO_2 emissions if it is assumed that increased biomass usage in the biorefinery does not lead to increased emissions elsewhere in the surrounding system. As can be seen, all the technologies investigated now show a considerable potential to reduce CO_2 emissions, in line with the results shown in the EU study. When the emissions associated with the alternative use of biomass are not considered, the main differences between this study and the EU study are that emissions associated with marginal fossil fuel-based electricity generation are considered instead of recalculating electricity to biomass, and that surplus heat is assumed to be used for district heating. Again the BLG process with its large electricity deficit shows the largest variation when the reference electricity production is varied, followed by the BIGCC process, due to its large district heat delivery. It should be noted that in this case there is no CO_2 emission credit for the indirect contribution to a decreased use of biomass, resulting from delivery of excess heat to the district heating system.

The BIGCC case where the electricity is used to charge battery-powered vehicles is the only studied case that consistently shows a potential for CO_2 reduction. The main reason for the good CO_2 performance is the high vehicle efficiency of the BPEV, compared to the other vehicle powertrains considered. However, BPEVs are primarily an option for personal transportation, not for heavy vehicles, and can thus only be expected to cover a part of the transportation need. Therefore, comparison of biomass-based motor fuels with fossil-based ones, for usage in for example hybrid vehicles, should be interesting.

As described in Chapter 5, only oil-based motor fuels are considered as alternative transportation technologies in the other papers. As can be seen from Table 3, coal-based motor fuels more than double the CO_2 emissions from the transport sector and this is considered less probable. However, if CCS is considered in connection with motor fuel production via coal gasification, the emissions are much closer to the emissions for oil-based motor fuels.

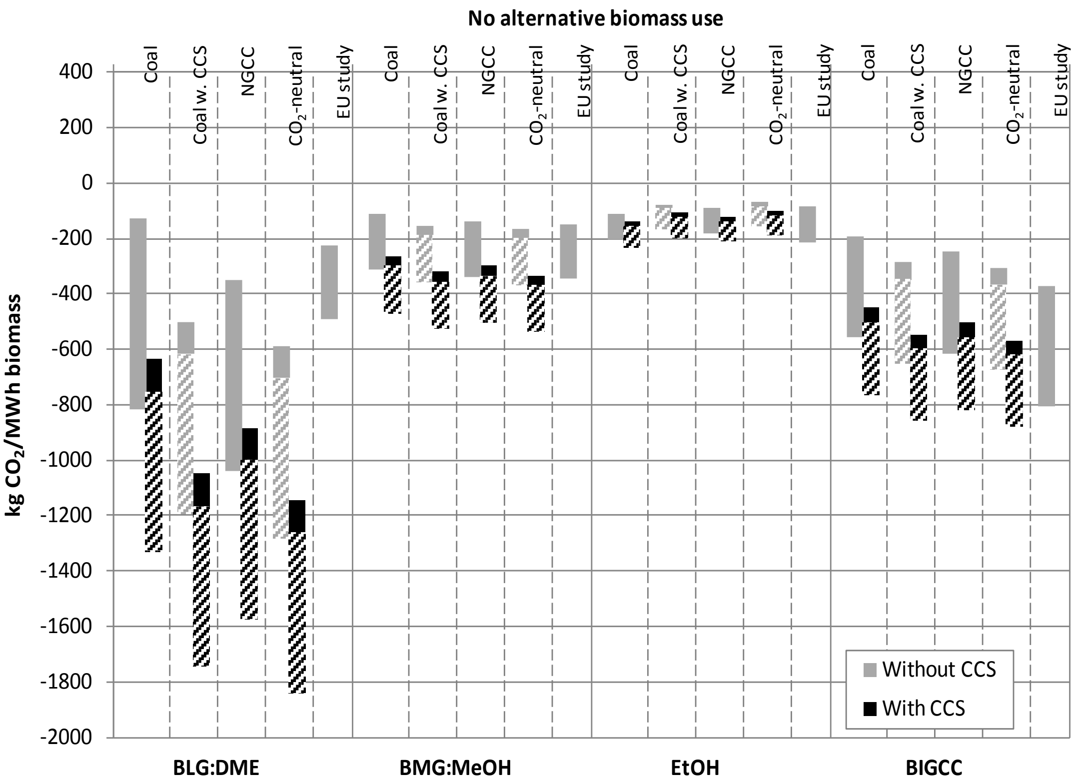

Figure 10. *Net CO_2 emissions for the studied cases when no alternative biomass use is assumed, for four different reference electricity technologies over a range representing the three marginal transportation technologies. Striped bars indicate combinations that are considered less probable. The results from the EU study, recalculated into CO_2 emissions per unit of biomass for the three marginal transportation technologies, are included for comparison.*

6.1.5 Conclusions

The results from Paper I show that it is important to take account of the fact that biomass and land for biomass production will be limited resources in the near future. Failure to expand the system to take alternative use of the biomass feedstock into account may result in overestimation of the potential of biomass-based transportation to contribute to reduced CO_2 emissions. This is particularly important when evaluating technologies that are expected to use a substantial amount of the available biomass in the future, as is the case with many biofuel technologies and other types of biorefinery concepts. Furthermore, it has been shown that when the assumptions about surrounding systems are varied, very different values regarding the potential to reduce CO_2 emissions by using biomass-based transportation are obtained.

6.2 Paper IV

This section summarizes Paper IV, "The effect of increased heat integration on the cost for producing DME via black liquor gasification". The economic analysis has been updated and inclusion of the effect on the global CO_2 emissions has been added, compared to the appended paper. Furthermore, a comparison with the mill reference case incorporating increased heat integration has been added.

6.2.1 Aim and procedure

The aim of this paper was to investigate the effect of increased heat integration on the net annual profit (NAP) and CO_2 emissions from a biorefinery plant producing DME via black liquor gasification. The study is based on the study by Ekbom et al. (2005), where integration of a BLGMF plant producing DME with a future kraft market pulp mill (steam use of approximately 11 GJ/ADt) was considered. High-temperature excess heat in the BLGMF plant is used to produce steam, partly used internally at the BLGMF plant, partly used at the mill. Additional steam for the mill is produced in a biomass fired CHP plant. Low-temperature excess heat from the BLGMF plant is used in the mill's secondary heat system, together with low-temperature excess heat from the mill. In this study, the secondary heat system is redesigned in order to make more heat available at sufficiently high temperature levels that it can be used to partly replace utility steam in the evaporation plant of the mill. As a result of lower mill steam demand, the size of the CHP plant is reduced and consequently the biomass use decreases. At the same time, however, the need for external electricity increases. The investment costs for the studied steam-saving measures are estimated and the effects on NAP and global CO_2 emissions are shown.

6.2.2 Mass and energy balances and investment costs

Table 4 shows the mass and energy balances and investment costs for the mill with BLGMF with increased heat integration (IHI) in comparison to the BLGMF base case and the mill reference case with a recovery boiler (RB). For details about the steam-saving measures, see Paper IV. In the mill reference case, there is a substantial steam surplus that is used to produce additional electricity. In the BLGMF base case, there is a substantial steam deficit that is covered by a biomass-fired CHP plant. Table 4 also includes a case where increased heat integration is considered for the mill with a recovery boiler. It has been assumed that the same amount of steam could be saved at the same cost. This corresponds to an overestimation of the steam-saving potential, or alternatively underestimating the cost to achieve the same steam saving. However, the absolute majority of streams involved in the calculations originate from the mill processes and not the BLGMF plant, and are thus the same in the case of a RB as in the case of a BLGMF plant. From Table 4 one can see that the use of bark (or other wood fuels) in the CHP plant is significantly reduced, from 143 to 66 MW, if increased heat integration is considered for the BLGMF case. However, this further decreases the electricity production, from 43 to 25 MW, and the net usage of electricity compared to the mill reference case further increases. If increased heat integration is considered in the RB case, this leads to increased electricity production, from 104 to 117 MW (the mill steam surplus increases and thus the production of condensing power). Thus, if the BLGMF case with increased heat integration is compared with the RB case with increased heat integration, the net usage of electricity increases even further.

Table 4. *Mass and energy balances and investment costs for the mill with BLGMF with increased heat integration in comparison to the BLGMF base case and the mill reference case with a RB. Furthermore, the case with increased heat integration for the mill with RB is shown.*

		RB ref. case	**BLGMF base case**	**BLGMF with IHI**	**RB with IHI**
Black liquor	MW	487	487	487	487
DME	MW	-	275	275	-
Electricity					
Production	MW	104	43	25	117
Consumption	MW	59	99	99	59
Surplus/Deficit	MW	45	-55	-74	58
Bark/Wood fuel					
Mill excess	MW	32	18[a]	18[a]	32
Consumption in CHP plant	MW	-	143	66	-
Surplus/Deficit	MW	32	-125	-48	32
Total investment cost	M€	210	443	420	225

[a] Larger bark use in the lime kiln due to different green liquor composition in the BLGMF case compared to the RB case.

The total investment cost is more than twice as high for the investment in a BLGMF plant compared to the investment in a new RB. By considering increased heat integration in the BLGMF case, the investment cost decreases, from 443 to 420 M€. The investment cost for the CHP plant is decreased by 34 M€ and the investment cost for the evaporation plant and the secondary heat system is increased by 11 M€, resulting in a total decrease of the additional investment cost of 23 M€. The investment cost for the RB case increases for the case of increased heat integration; 11 M€ for the steam-saving measures and 4 M€ incremental cost for the condensing steam turbine. Thus, the incremental investment cost for BLGMF compared to RB decreases from 233 M€ (443 compared to 210) to 195 M€ (420 compared to 225), which corresponds to 16%, if increased heat integration is considered. The annual operating and maintenance costs are assumed to be 9 M€ higher for the BLGMF cases compared to the RB cases.

6.2.3 Economic performance and CO_2 emission balances

The NAP and CO_2 emission balances are calculated using the different future energy market scenarios presented in Table 1 (Section 5.1).

The results concerning NAP are presented in Figure 11. Investments in increased heat integration are profitable for the RB case, as can be seen from the first groups of bars in Figure 11. This is because the increased revenue from sold electricity is greater than the increased capital cost. Δ NAP is highly positive, if investment in a BLGMF plant is compared with investment in a new RB. Thus, the revenue from sold DME is significantly higher than the increased costs/decreased revenues for capital, electricity and wood fuel. The profit could be further increased if increased heat integration is considered (third groups of bars). Thus, the decreased cost for wood fuel and capital is greater than the decreased revenue from sold electricity. A more fair comparison is to relate the case where increased heat integration is considered for the BLGMF case with the corresponding RB case (fourth group of bars). However, the difference in NAP between the BLGMF and RB cases is higher if increased heat integration is considered (compare the fourth group of bars with the second). This is because NAP increases more when increased heat integration is considered for the BLGMF case compared to the RB case (compare the fifth group of bars with the first).

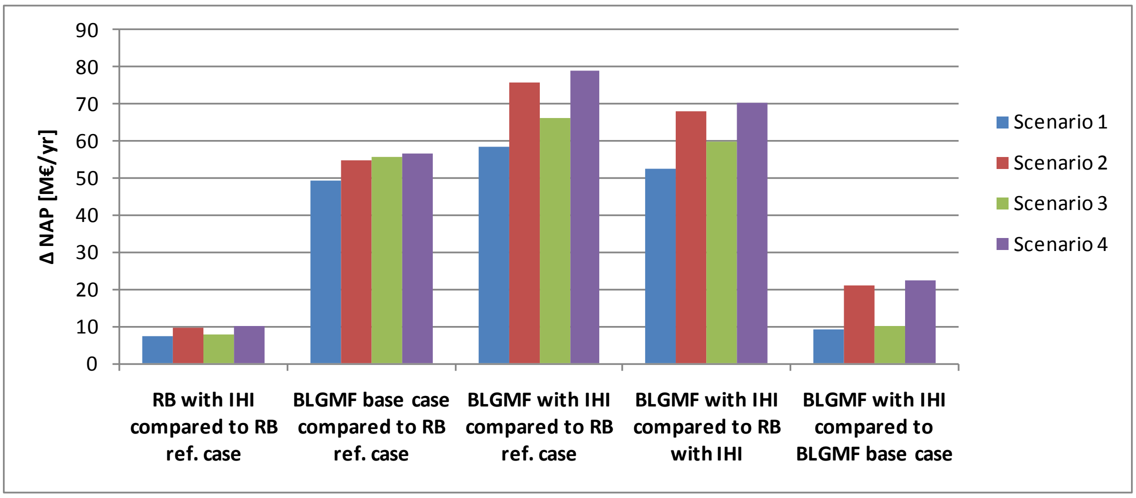

Figure 11. *Results concerning net annual profit (NAP).*

Some differences between the different scenarios can be seen. In Scenarios 1 and 3, the ratio between the prices for wood fuel and electricity (including green electricity policy instrument) is lower than in Scenarios 2 and 4 (0.35 compared to 0.5). Thus, the steam-saving measures in the BLGMF case are less profitable in Scenarios 1 and 3 compared to Scenarios 2 and 4 since the resulting effect is decreased usage of wood fuel but also decreased production of electricity. As can be seen, the increase of NAP as a result of increased heat integration is only slightly higher for the BLGMF case compared to the RB case in Scenarios 1 and 3. However, in Scenarios 2 and 4, the increase of NAP is significantly higher for the BLGMF case compared to the RB case.

Figure 12 shows the results concerning global CO_2 emissions. Negative values correspond to decreased CO_2 emissions. Investment in steam-saving measures in the RB case lead to decreased CO_2 emissions, since the resulting increased electricity generation can avoid electricity generation in coal power plants. The effect is significantly lower in Scenarios 2 and 4, where the coal power plants are assumed to be equipped with CCS. Investing in a BLGMF plant instead of a new RB leads to increased emissions in Scenarios 1 and 3. Thus, the decrease of CO_2 emissions in the transport sector is lower than the increased emissions in the power sector and the increased emissions elsewhere in the system resulting from the increased use of wood fuel at the mill. However, in Scenarios 2 and 4, the CO_2 emissions decrease as a result of significantly lower increase of emissions in the power sector.

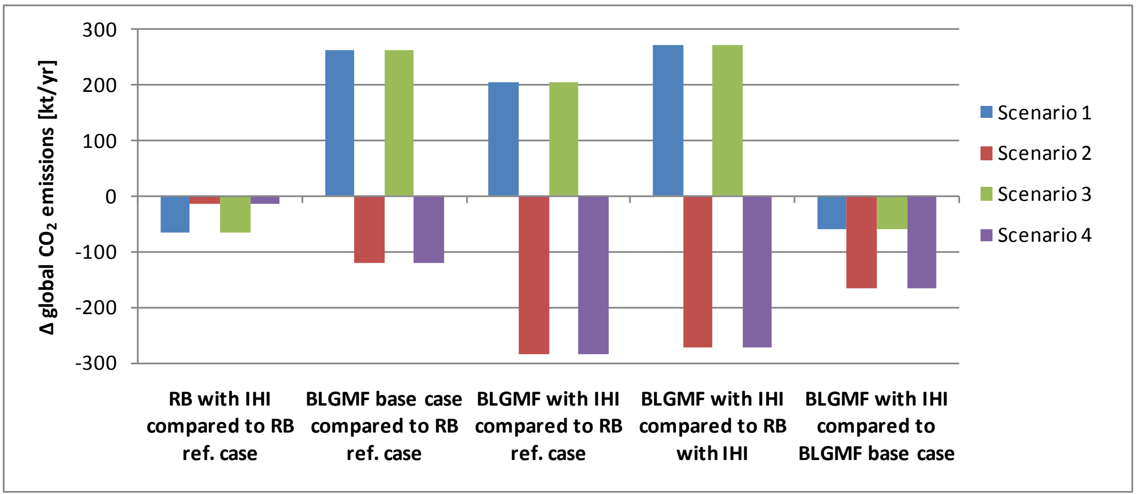

Figure 12. *Results concerning global CO_2 emissions.*

Increased heat integration improves the CO_2 emissions balances if it is considered for the BLGMF case compared to if it is not considered. However, as discussed, the BLGMF case with increased heat integration should be compared to the corresponding RB case. This comparison indicates that the global CO_2 emissions are somewhat further increased in Scenarios 1 and 3. However, the increase is insignificant. In Scenarios 2 and 4, however, the decrease of CO_2 emissions is much higher in the BLGMF case than in the RB case, since the marginal electricity production technology is coal with CCS, thereby decreasing the reduction in the RB case and increasing the reduction in the BLGMF case.

6.2.4 Conclusions

This paper investigated the effect of increased heat integration on the profitability and CO_2 emissions effect for producing DME via black liquor gasification. The results concerning profitability show that if a future market pulp mill invests in a BLGMF plant producing DME, the net annual profit is significantly higher compared to investing in a new recovery boiler. The effect on global CO_2 emissions varies from a significant increase to a significant decrease depending on the assumed marginal electricity production technology. If increased heat integration by using excess heat to replace steam in the evaporation plant is considered, both for the mill with BLGMF and RB technology, the following conclusions can be drawn:

- The net usage of wood fuel in the BLGMF case, compared to the RB case, decreases by 49%, whereas the net usage of electricity increases by 32%.
- The incremental investment cost for BLGMF, compared to RB, decreases by 16%.
- Investments in increased heat integration are profitable for both the RB and BLGMF cases. However, it is more profitable for the BLGMF case, especially in scenarios with a relatively high ratio between the biomass and electricity price. The net annual profit of an investment in a BLGMF plant, compared to investing in a new recovery boiler, is increased by 7% in scenarios with a relatively low ratio between the biomass and electricity price, and by 24% in scenarios with a relatively high price ratio.
- Investments in increased heat integration improve the CO_2 emissions balances for both the RB and BLGMF cases. For which case the improvement is greatest is dependent on the marginal electricity production technology that is assumed. The effect on global CO_2 emissions resulting from an investment in a BLGMF plant, instead of investing in a new recovery boiler, is somewhat further increased (3%) in scenarios with coal power as marginal electricity production technology. However, in scenarios with coal power with CCS as marginal electricity production technology, the CO_2 emissions resulting from an investment in a BLGMF plant are further decreased (129%) compared to investing in a new recovery boiler.

6.3 Paper V

This section summarizes Paper V, "Comparison of black liquor gasification with other pulping biorefinery concepts – Systems analysis of economic performance and CO_2 emissions".

6.3.1 Aim and procedure

This study presents the results of a systems analysis in which black liquor gasification is compared with other recovery boiler-based biorefinery concepts from economic and climatic points of view. First, mass and energy balances for the integration of different concepts are calculated, and thereafter the economic performance and CO_2 emission balances are calculated for different future energy market conditions. The study includes different types of mills, with different steam requirements, for which the applicability and performance of various biorefinery concepts vary.

6.3.2 The studied mill system

The studied system consists of a pulp mill that is evaluating options for replacement of an old recovery boiler. Available investment options include: (1) a new recovery boiler, (2) a BLGCC plant or (3) a BLGMF plant. Three different steam demands are considered for the mill: 10, 14.5 and 19 GJ/ADt. Both for the recovery boiler and for the BLG options, it is assumed to be possible to capture CO_2 and send it to storage. It is also assumed to be possible to extract lignin from the black liquor[3]. Depending on the mill steam demand and the characteristics of the recovery boiler or the BLG case, the mill will have a surplus or deficit of steam. A surplus of steam is assumed to be used for electricity (El) production in a condensing steam turbine. A steam deficit is assumed to be handled by firing falling bark and purchased bark or other wood fuels, if required, in a bark boiler (BB) connected to a steam turbine – or by gasifying wood fuel for production of either electricity and steam in a gas turbine combined cycle (BIGCC), or motor fuels, electricity and steam (BIGMF and BIGCC). For both the bark boiler and the solid biomass gasification options, it is assumed that it is possible to capture CO_2. The equipment assumed to be required for balancing a steam deficit, or steam surplus, is also assumed to be in need of investments. Figure 13 presents the studied mill system.

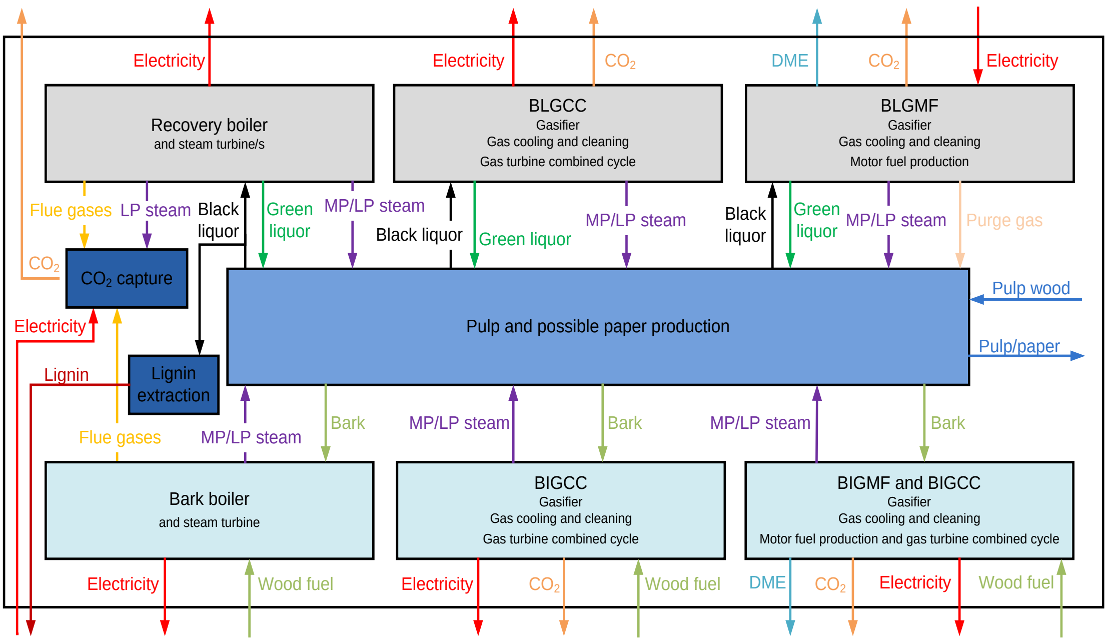

Figure 13. *A schematic representation of the main energy and material streams for the studied mill and different integrated biorefinery process concepts.*

In order to limit the number of cases, some combinations that are considered less probable are excluded (see Paper V). For a more detailed description of the different mill processes, including mass and energy balances, investment costs, and operation and maintenance costs, see Paper V.

[3] This is however not considered for the BLG options.

6.3.3 Studied cases

Tables 5-7 present the mass and energy balances, investment cost and operation and maintenance (O&M) cost for the studied investment options. The cases are named by first specifying the energy and chemical recovery technology considered, that is RB/BLGCC/BLGMF, and whether lignin extraction (LE) and/or CCS is included (left side of the ":"). Thereafter follows an abbreviation indicating the technology considered to balance the plant's steam demand, that is El/BB/BIGCC/BIGMF and BIGCC[4], and whether CCS is included (right side of the ":"). Cases that include CCS are indicated in bold type, and cases that have a net deficit of wood fuel (WF) above 500 MW are underlined. Negative values indicate a net deficit.

For the mill steam demand of 10 GJ/ADt, the mill reference case (case 1) has a substantial surplus of steam that is used to produce additional electricity. None of the cases has a net wood fuel deficit above 500 MW for this mill steam demand.

In the reference case (case 2) for the mill steam demand of 14.5 GJ/ADt, the steam production in the recovery boiler is equal to the mill steam use. This implies that all other cases have a steam deficit. Note that the net deficit of wood fuel exceeds 500 MW for several of the cases. Since there is no steam surplus in the reference case, the steam deficit and thus the need for extra biomass are larger for the other cases than if the mill reference case has a steam surplus, as is the case for 10 GJ/ADt.

The reference case (case 3) for the mill steam demand of 19 GJ/ADt has a substantial deficit of steam that is covered by firing falling bark and purchased wood fuel in a boiler connected to a steam turbine. The net deficit of wood fuel is above 500 MW for most of the other cases. It is difficult to draw general conclusions about the amount of wood fuel that can be imported to a mill, and the upper limit depends on a number of factors, particularly the size of the collection area that has to be considered in order to gather the required amount of wood fuel. In Tables 6 and 7, cases that have to purchase more than 500 MW of wood fuel are underlined. This can be related to the energy content in the incoming pulp wood for this mill size, which is around 700 MW. It can also be related to the estimated possible increase in future supply of biomass, compared to the current level for Sweden where many kraft pulp mills are located. Several studies estimate a possible increase of up to approximately 400 PJ/year (~13 GW) (Lindfeldt et al., 2010). Not only is the need for external wood fuel very large in several cases for the mill steam demands of 14.5 and 19 GJ/ADt. So are the investment costs, as can be seen in Tables 6 and 7. For a discussion regarding uncertainties and improvement potential for the studied cases, see Paper V.

[4] Referred to only as BIGMF here.

Table 5. *Mass and energy balances, investment cost and operation and maintenance cost for the studied investment options for the mill steam demand of ≈10 GJ/ADt.*

Case No	Case	MF [MW]	El [MW]	WF [MW]	Lignin [MW]	CO_2[1] [t/h]	Inv. cost [M€]	O&M cost [M€/yr]
1 - Ref. case	RB:El	-	35	32	-	-	258	9
8	RBLE:El	-	-7	32	104	-	240	15
14	**RBCCS:BBCCS**	**-**	**-3**	**14**	**-**	**143**	**334**	**20**
17	**RBLECCS:BBCCS**	**-**	**10**	**-142**	**104**	**164**	**455**	**33**
18	**RBLECCS:BIGCCCCS**	**-**	**39**	**-225**	**104**	**168**	**552**	**36**
19	**RBLECCS:BIGMFCCS**	**124**	**6**	**-337**	**104**	**169**	**611**	**38**
20	BLGCC:El	-	68	26	-	-	354	12
23	**BLGCCCCS:El**	**-**	**29**	**26**	**-**	**122**	**390**	**21**
27	BLGMF:BB	268	-99	-36	-	-	416	14
28	BLGMF:BIGCC	268	8	-157	-	-	592	20
29	BLGMF:BIGMF	370	-21	-278	-	-	603	20
30	**BLGMFCCS:BB**	**268**	**-109**	**-36**	**-**	**82**	**423**	**19**
31	**BLGMFCCS:BBCCS**	**268**	**-95**	**-129**	**-**	**147**	**529**	**27**
32	**BLGMFCCS:BIGCCCCS**	**268**	**-17**	**-173**	**-**	**128**	**580**	**27**
33	**BLGMFCCS:BIGMFCCS**	**370**	**-38**	**-278**	**-**	**132**	**612**	**29**

[1] Captured CO_2.

Table 6. *Mass and energy balances, investment cost and operation and maintenance cost for the studied investment options for the mill steam demand of ≈14.5 GJ/ADt.*

Case No	Case	MF [MW]	El [MW]	WF [MW]	Lignin [MW]	CO_2[1] [t/h]	Inv. cost [M€]	O&M cost [M€/yr]
2 - Ref. case	RB	-	10	32	-	-	233	8
9	RBLE:BB	-	12	-96	104	-	321	18
10	RBLE:BIGCC	-	93	-276	104	-	462	23
11	**RBLE:BIGCCCCS**	**-**	**71**	**-303**	**104**	**78**	**521**	**30**
12	RBLE:BIGMF	161	39	-445	104	-	583	27
13	**RBLE:BIGMFCCS**	**161**	**28**	**-445**	**104**	**79**	**590**	**32**
14	**RBCCS:BBCCS**	**-**	**51**	**-302**	**-**	**244**	**563**	**27**
15	**RBCCS:BIGCCCCS**	**-**	**120**	**-459**	**-**	**252**	**707**	**39**
16	**RBCCS:BIGMFCCS**	**242**	**59**	**-684**	**-**	**255**	**805**	**43**
17	**RBLECCS:BBCCS**	**-**	**56**	**-458**	**104**	**265**	**626**	**45**
18	**RBLECCS:BIGCCCCS**	**-**	**158**	**-689**	**104**	**276**	**816**	**52**
19	**RBLECCS:BIGMFCCS**	**361**	**66**	**-1038**	**104**	**313**	**951**	**59**
21	BLGCC:BB	-	71	-59	-	-	407	14
22	BLGCC:BIGCC	-	125	-177	-	-	473	16
24	**BLGCCCCS:BB**	**-**	**38**	**-73**	**-**	**122**	**454**	**23**
25	**BLGCCCCS:BBCCS**	**-**	**47**	**-150**	**-**	**178**	**542**	**29**
26	**BLGCCCCS:BIGCCCCS**	**-**	**84**	**-233**	**-**	**182**	**576**	**31**
27	BLGMF:BB	268	-55	-219	-	-	509	17
28	BLGMF:BIGCC	268	109	-581	-	-	727	24
29	BLGMF:BIGMF	608	11	-984	-	-	905	30
30	**BLGMFCCS:BB**	**268**	**-66**	**-219**	**-**	**82**	**516**	**22**
31	**BLGMFCCS:BBCCS**	**268**	**-33**	**-441**	**-**	**247**	**713**	**39**
32	**BLGMFCCS:BIGCCCCS**	**268**	**56**	**-634**	**-**	**236**	**822**	**42**
33	**BLGMFCCS:BIGMFCCS**	**608**	**-21**	**-984**	**-**	**248**	**916**	**46**

[1] Captured CO_2.

Table 7. *Mass and energy balances, investment cost and operation and maintenance cost for the studied investment options for the mill steam demand of ≈19 GJ/ADt.*

Case No	**Case**	**MF [MW]**	**El [MW]**	**WF [MW]**	**Lignin [MW]**	**CO_2[1] [t/h]**	**Inv. cost [M€]**	**O&M cost [M€/yr]**
3 - Ref. case	RB:BB	-	45	-140	-	-	348	12
4	RB:BIGCC	-	155	-382	-	-	520	17
5	**RB:BIGCCCCS**	-	**125**	**-417**	-	**105**	**591**	**20**
6	RB:BIGMF	217	82	-612	-	-	669	22
7	**RB:BIGMFCCS**	**217**	**68**	**-612**	-	**106**	**677**	**23**
9	RBLE:BB	-	47	-268	104	-	399	21
10	RBLE:BIGCC	-	240	-690	104	-	680	30
11	**RBLE:BIGCCCCS**	-	**188**	**-753**	**104**	**183**	**753**	**32**
12	RBLE:BIGMF	391	117	-1126	104	-	880	37
13	**RBLE:BIGMFCCS**	**391**	**92**	**-1126**	**104**	**191**	**893**	**37**
14	**RBCCS:BBCCS**	-	**103**	**-608**	-	**342**	**710**	**45**
15	**RBCCS:BIGCCCCS**	-	**253**	**-908**	-	**357**	**920**	**53**
16	**RBCCS:BIGMFCCS**	**471**	**139**	**-1365**	-	**367**	**1081**	**59**
17	**RBLECCS:BBCCS**	-	**98**	**-765**	**104**	**363**	**762**	**55**
18	**RBLECCS:BIGCCCCS**	-	**289**	**-1139**	**104**	**381**	**1010**	**65**
19	**RBLECCS:BIGMFCCS**	**590**	**148**	**-1718**	**104**	**396**	**1202**	**72**
21	BLGCC:BB	-	106	-228	-	-	491	16
22	BLGCC:BIGCC	-	268	-585	-	-	657	22
24	**BLGCCCCS:BB**	-	**73**	**-243**	-	**122**	**536**	**25**
25	**BLGCCCCS:BBCCS**	-	**99**	**-452**	-	**274**	**712**	**41**
26	**BLGCCCCS:BIGCCCCS**	-	**198**	**-677**	-	**286**	**804**	**45**
27	BLGMF:BB	268	-15	-395	-	-	581	19
28	BLGMF:BIGCC	268	252	-989	-	-	887	30
29	BLGMF:BIGMF	838	87	-1665	-	-	1154	38
30	**BLGMFCCS:BB**	**268**	**-25**	**-395**	-	**82**	**588**	**25**
31	**BLGMFCCS:BBCCS**	**268**	**34**	**-786**	-	**357**	**878**	**51**
32	**BLGMFCCS:BIGCCCCS**	**268**	**169**	**-1077**	-	**339**	**1020**	**55**
33	**BLGMFCCS:BIGMFCCS**	**838**	**41**	**-1665**	-	**360**	**1174**	**62**

[1] Captured CO_2.

6.3.4 Economic performance and CO_2 emission balances

The NAP and global CO_2 emission balances are calculated for the studied cases using the different future energy market scenarios presented in Table 1 (Section 5.1).

To further study the influence of policy instruments, a sensitivity analysis where the policy instrument for green electricity is removed has been included. Table 8 shows the effect on the wood fuel price and the DME policy instrument when the policy instrument for green electricity is removed. In Scenarios 3 and 4, removal of the policy instrument for green electricity results in the DME plant having a slightly higher WTP for wood fuel than the coal power plant.

Table 8. *The effect on the wood fuel price and the DME policy instrument when the policy instrument for green electricity is removed.*

Scenario input		**1**	**2**	**3**	**4**
Fossil fuel price level		Low	Low	High	High
CO_2 emission charge		Low	High	Low	High
Green electricity policy instrument	[€/MWh]	0	0	0	0
Resulting values of prices and policy instruments [€/MWh]					
Bark/by-products/wood chips		14	39	21	46
DME policy instrument		11	15	0	0

For the technologies not yet commercial today, there is a large uncertainty when it comes to investment costs, as discussed in Section 4.8. Therefore, as a sensitivity analysis, the investment costs for non-commercial technologies are increased by 30%. As a third case, a sensitivity analysis combining removed policy instrument for green electricity and increased investment cost for non-commercial technologies is included.

If it is assumed that coal power plants and DME plants are still both marginal users of wood fuel (which should be a reasonable assumption; the DME plants' WTP is only slightly higher in Scenarios 3 and 4), the CO_2 emission values are the same as for the scenario base case (see Table 1) and thus only NAP is influenced by these sensitivity analysis cases.

Figures 14-16 present the results for the studied cases for the three different mill steam demand levels and four different future energy market scenarios considered. The net annual profit and global CO_2 emissions for each case are calculated relative to the mill reference case. The mill reference case is thus represented by the intersection of the x-axis and y-axis (left diagrams), and the changes between the mill reference case values and the different case values are thus shown on the x-axis and the y-axis (Δ NAP and Δ global CO_2 emissions). Cases that are positioned in the lower right quadrant have both higher NAP and lower global CO_2 emissions than the mill reference case.

RB-based cases are green, BLGCC-based cases are blue and BLGMF-based cases are orange. Cases that include CCS are filled, and cases that have a net deficit of wood fuel above 500 MW are underlined.

The right diagrams present the results from the sensitivity analysis and show how Δ NAP is influenced by removed support for green electricity (a), increase of investment costs for non-commercial technologies by 30% (b), and removed support for green electricity and increase of investment costs for non-commercial technologies by 30% (c).

It is debatable whether there will be a widespread infrastructure for storing the CO_2 if capture is not widely introduced in the power sector with its very large emission point sources, as discussed in Section 4.5. Therefore, CCS is not considered as an option in Scenarios 1 and 3.

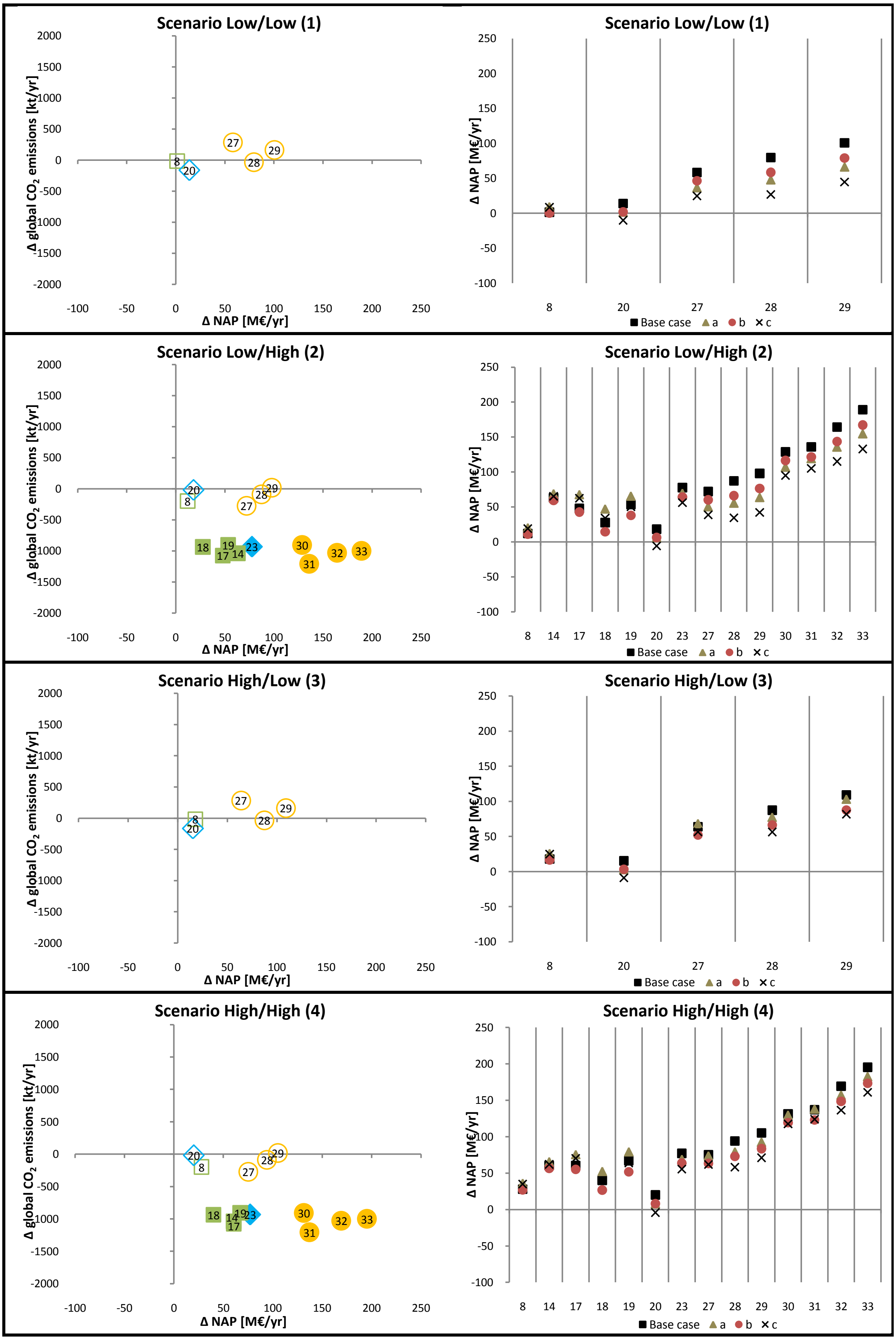

Figure 14. *Results for the studied cases for the mill steam demand of ≈10 GJ/ADt presented in Table 5, under the different energy market scenarios presented in Table 1.*

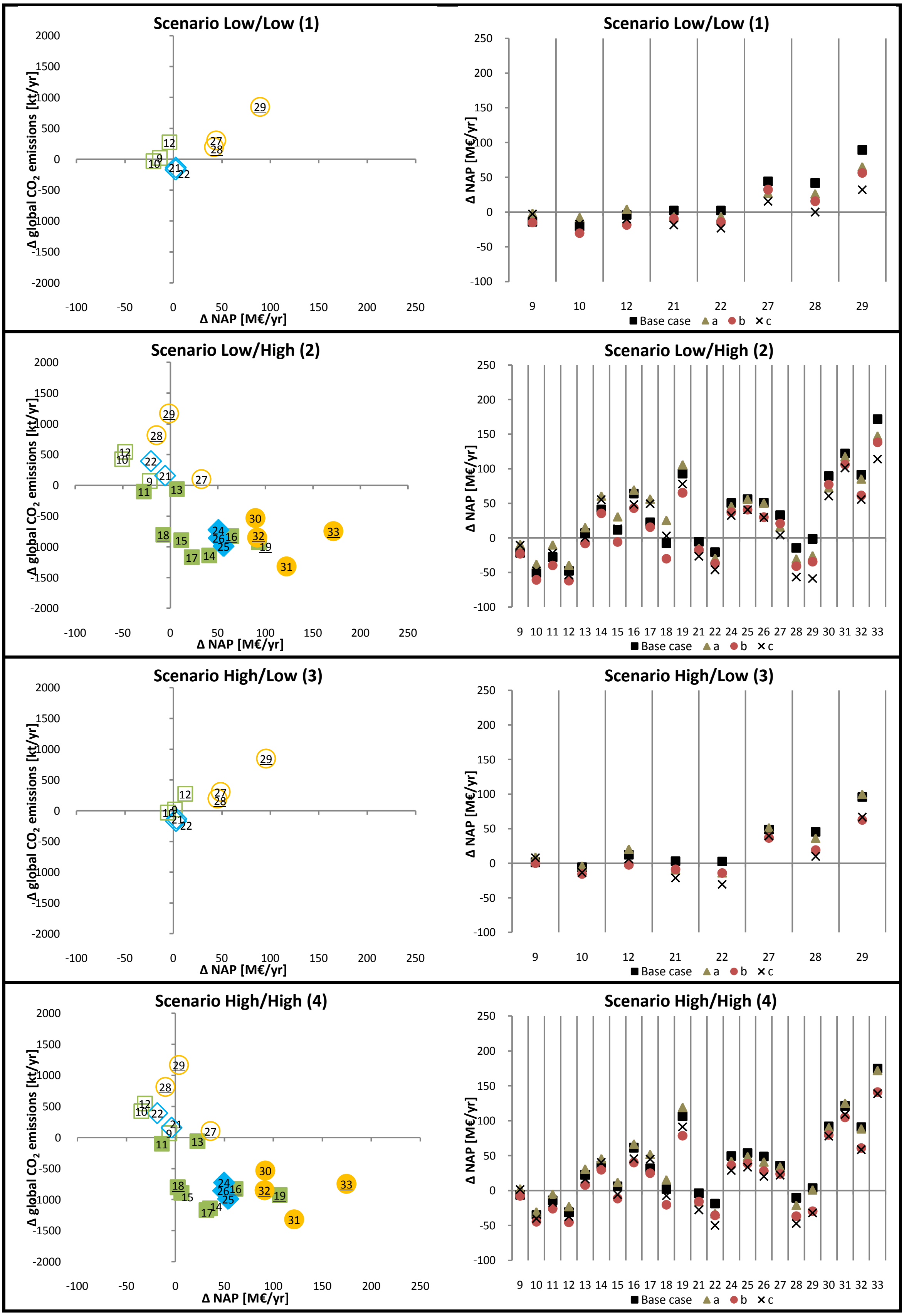

Figure 15. *Results for the studied cases for the mill steam demand of ≈14.5 GJ/ADt presented in Table 6, under the different energy market scenarios presented in Table 1.*

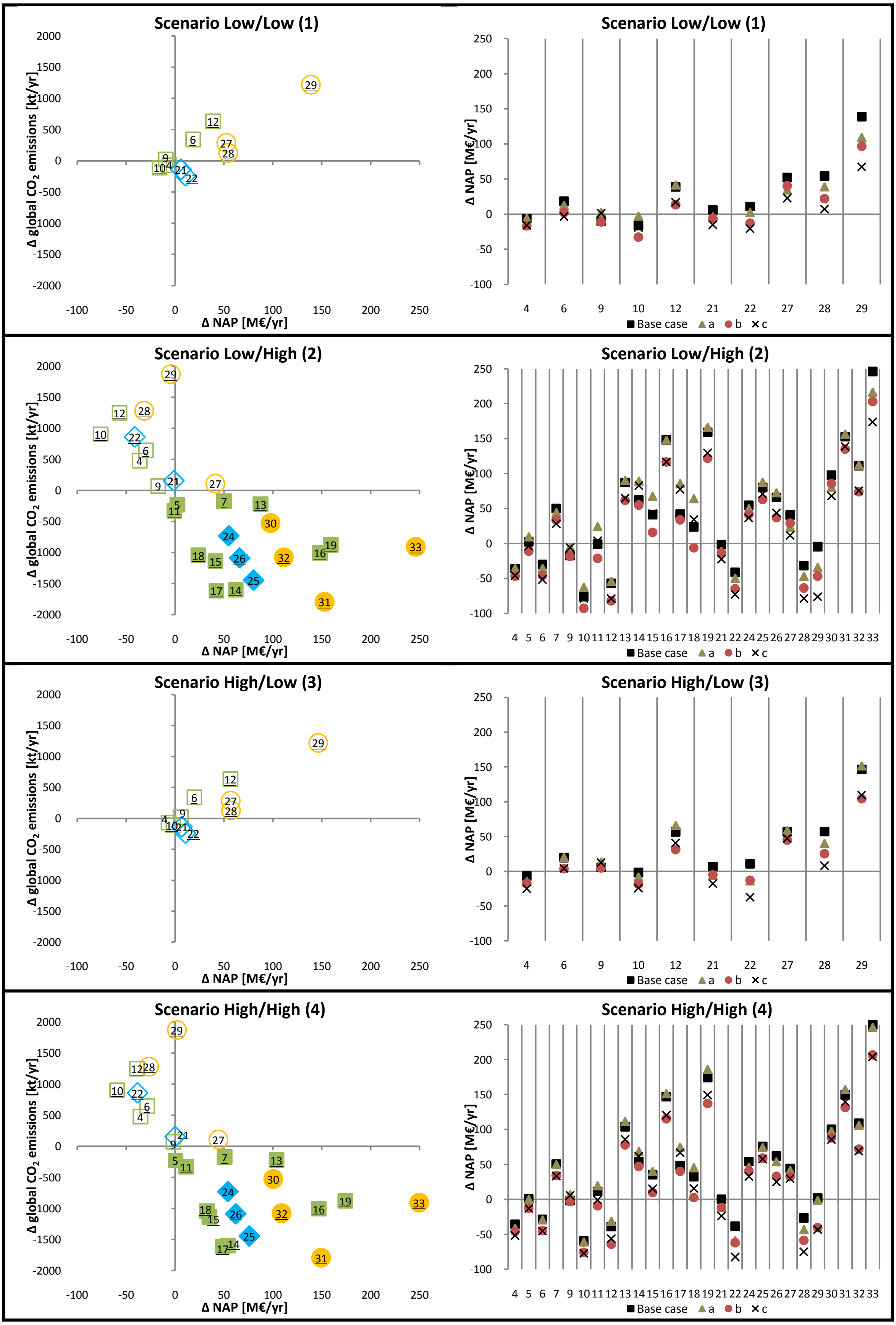

Figure 16. *Results for the studied cases for the mill steam demand of ≈19 GJ/ADt presented in Table 7, under the different energy market scenarios presented in Table 1.*

6.3.4.1 The mill steam demand of 10 GJ/ADt

The BLGMF cases without CCS (cases 27-29) show a relatively consistent positive Δ NAP over the scenarios. Case 28 (BLGMF:BIGCC) has a better economic performance than case 27 (BLGMF:BB); the economic value of the decreased net use of electricity is larger than the increased costs for wood fuel and capital. Case 29 (BLGMF:BIGMF) also has a better economic performance than case 27 (and 28); the profit from the decreased net use of electricity and increased production of DME is larger than the increased costs for wood fuel and capital.

The CO_2 effect for cases 27-29 varies between the scenarios. In Scenarios 1 and 3, cases 27 and 29 have a net CO_2 emissions increase. In Scenarios 2 and 4 the marginal electricity production changes from coal power to coal power with CCS, resulting in a net CO_2 emission decrease for case 27 and a neutral CO_2 balance for case 29.

In the scenarios with a high CO_2 charge (Scenarios 2 and 4 where CCS is considered), the BLGMF cases including CCS (cases 30-33) have a much better economic performance than the cases without CCS (cases 27-29). Also the potential to reduce global CO_2 emissions is much higher with CCS than the cases without CCS.

Case 20 (BLGCC:El) has a consistent positive Δ NAP in all scenarios. The increased production of electricity generates higher revenue compared to the increased capital cost (and small decreased revenue from selling wood fuel). The CO_2 reduction potential is greater in Scenarios 1 and 3, since the increased electricity produced replaces electricity from coal power in these scenarios, compared to Scenarios 2 and 4 where electricity from coal power plants equipped with CCS is replaced. As for the BLGMF cases, inclusion of CCS in Scenarios 2 and 4 significantly improves both the economic performance and the CO_2 reduction potential (case 23).

Case 8 (RBLE:El) has a positive Δ NAP that increases over the scenarios since the ratio between the lignin price and the electricity price increases (the case has a net use of electricity compared to the mill reference case). It shows a potential to reduce CO_2 emissions in all scenarios. In Scenarios 2 and 4, the RB cases with CCS (14, 17-19) all achieve a clear positive Δ NAP and a high CO_2 reduction potential.

6.3.4.2 The mill steam demands of 14.5 and 19 GJ/ADt

Figure 17 shows how Δ NAP varies with the mill steam demand level for case 27 (BLGMF:BB), case 20/21 (BLGCC:El/BLGCC:BB) and case 8/9 (RBLE:El/RBLE:BB). As can be seen from the figure, Δ NAP is largest for the mill steam demand of 10 GJ/ADt and has a minimum at 14.5 GJ/ADt after which it starts to increase. This is because the increased costs/decreased revenues compared to the mill reference case are highest at 14.5 and lowest at 10 GJ/ADt.

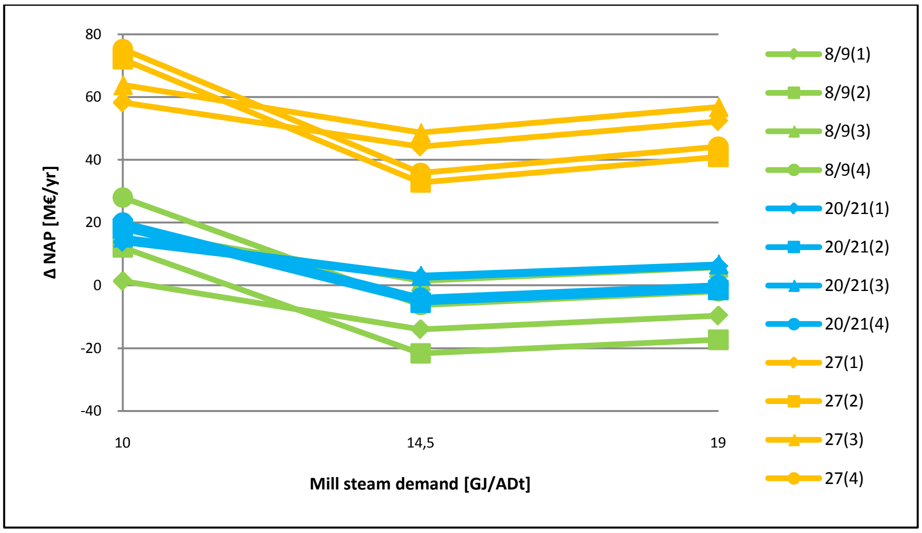

Figure 17. *Δ NAP for the four energy market scenarios presented in Table 1 as a function of mill steam demand level for cases 8/9, 20/21 and 27.*

Take case 27 as an example. For 10 GJ/ADt, the mill reference case has a large steam surplus and produces considerable amounts of electricity in the condensing turbine unit. If the recovery boiler is replaced by a BLGMF plant, the large steam surplus becomes a steam deficit. Since there is a steam surplus in the reference case, the steam deficit and thus the need for extra wood fuel will not be as large as they would have been if the mill reference case had a steam deficit. However, the electricity balance changes considerably and the production decrease is very large in case 27 for the steam use of 10 GJ/ADt. Still, the sum of the decreased revenues/increased costs for wood fuel and electricity is lowest for this steam demand level. Also the incremental investment cost for the BLGMF case compared to the mill reference case is lowest at 10 GJ/ADt, since only a relatively small bark boiler is needed in the BLGMF case and a relatively expensive condensing turbine is used in the reference case. For 14.5 and 19 GJ/ADt, the sum of the decreased revenues/increased costs for wood fuel and electricity is approximately the same. Yet the incremental investment cost for case 27 compared to the mill reference case is lower for 19 compared to 14.5 GJ/ADt, and that is the reason for the difference in NAP between these two steam demand levels.

For 10 GJ/ADt, Δ NAP for case 27 is better in Scenarios 2 and 4 (see Figure 17) since increased costs for electricity and wood fuel are more than compensated by an increased revenue for DME. However, for 14.5 and 19 GJ/ADt, Δ NAP for case 27 is better in Scenarios 1 and 3 than in Scenarios 2 and 4. This is due to the changed energy balance; the net usage of electricity is lower but the net usage of wood fuel is much higher. This makes the increase of costs for wood fuel and electricity in Scenarios 2 and 4 higher for these steam demand levels compared to 10 GJ/ADt because the wood fuel price increases more in Scenarios 2 and 4 compared to the electricity price.

The decrease of Δ NAP for 14.5 and 19 GJ/ADt results in case 21 (BLGCC:BB) only being slightly profitable in Scenarios 1 and 3. Case 22 (BLGCC:BIGCC) has a Δ NAP similar to case 21 in Scenarios 1 and 3.

The net decrease of CO_2 emissions is smaller or the increase is greater for the cases presented in Figure 17, and for almost all the other cases without CCS as well (see Figures 15 and 16). Of the cases in Figure 17, only case 21 (BLGCC:BB) in Scenarios 1 and 3 shows a net decrease of CO_2 emissions for 14.5 and 19 GJ/ADt.

Case 28 (BLGMF:BIGCC) now has a poorer, or in some cases equal, economic performance compared to case 27 (BLGMF:BB). For case 29 (BLGMF:BIGMF), the increased costs for wood fuel and capital are lower than the decreased cost or increased revenue for electricity and the increased revenue for DME (compared to case 27) in Scenarios 1 and 3, but not in Scenarios 2 and 4. Both cases 28 and 29 have a net deficit of wood fuel greater than 500 MW for both 14.5 and 19 GJ/ADt.

Most of the RB cases without CCS generally have both a poor economic performance and CO_2 emissions reduction potential. In Scenarios 2 and 4, for example, they are all located in the upper left quadrant and thus have both a lower NAP and increased global CO_2 emissions compared to the mill reference cases. Several cases, for example case 4 (RB:BIGCC), are not profitable in any of the scenarios. Case 9 (RBLE:BB) has a slightly positive Δ NAP in Scenario 3, which has the highest ratio between the lignin and wood fuel price and thus benefits case 9.

Case 6 (RB:BIGMF) has a good economic performance in Scenarios 1 and 3, but not in Scenarios 2 and 4. However, case 7 (RB:BIGMFCCS) has a good economic performance in Scenarios 2 and 4. The BIGMF plant considered as marginal user of wood fuel is the same plant as considered for integration with the mill. The only difference is that the plant considered as marginal user of wood fuel is a stand-alone plant. This plant is assumed to send separated CO_2 to storage in the scenarios based on a high CO_2 charge, as described in Section 5.1. Thus, since the level of support needed is set based on a plant that sends CO_2 to storage in Scenarios 2 and 4, it is not strange that case 6 which does not include CCS has a poor economic performance in those scenarios. That cases 6 and 7 show a positive Δ NAP in Scenarios 1 and 3 and Scenarios 2 and 4, respectively, indicates the benefits of integrating the considered BIGMF plant with a pulp and paper mill compared to operating it in stand-alone mode.

As can be seen in Table 7, the external need for wood fuel in case 6 is high – 612 MW. However, the steam demand level of 19 GJ/ADt is representative of a future integrated fine paper mill with low energy efficiency, as discussed in Section 4.2. The level 14.5 GJ/ADt is representative of a future integrated fine paper mill with very high energy efficiency. It is therefore probable that future integrated fine paper mills will have a steam demand somewhere between these levels. Even if it is closer to

14.5 GJ/ADt, there will still be a significant steam deficit, and integration with for example a BIGMF plant could be considered. The need for external wood fuel would then be more reasonable than if the steam demand is 19 GJ/ADt. The BIGMF plant considered does not have a maximized DME yield and can be regarded as a mix of a BIGMF and BIGCC plant. The reason for choosing this plant is that it produces more steam per unit of wood fuel input than a corresponding plant with a maximized DME yield, and thus the size of the plant is smaller for a certain steam deficit. However, it could be interesting to consider a BIGMF plant with a higher DME yield if this does not lead to a too large external need for wood fuel.

All cases including CCS have a potential for reducing global CO_2 emissions and have significantly better economic performance than the corresponding cases without CCS.

6.3.4.3 The influence of policy instruments and increased investment costs

Δ NAP results for the BLGMF cases (27-33) are relatively sensitive to an increase of 30% for the investment costs (sensitivity analysis case *b*, Figures 14-16), since the investment costs are generally large. For 10 GJ/ADt, removed support for green electricity and subsequent decreased support for DME and lower wood fuel price (sensitivity analysis case *a*, Figures 14-16) lead to a significant decrease of Δ NAP in Scenarios 1 and 2, but in Scenarios 3 and 4 the decrease is less since the level of support for DME is lower in the base case for these scenarios. For 14.5 and 19 GJ/ADt, removed support for green electricity generally affects cases 27-33 negatively in Scenarios 1 and 2. This, for example, leads to only a slightly positive Δ NAP for case 27 in Scenario 2, if considered in combination with a higher investment cost (sensitivity analysis case *c*, Figures 14-16). For Scenarios 3 and 4, the effect of removed support for green electricity is only slightly negative or in some cases positive.

The BLGCC cases (20-25) are generally less affected by a 30% increase for investment costs, compared to the BLGMF cases. Removed support for green electricity slightly decreases Δ NAP for most of the BLGCC cases. However, in some cases with a large net use of wood fuel, Δ NAP increases slightly. If considered in combination with a higher investment cost, the BLGCC cases without CCS have a negative Δ NAP in all scenarios for all steam demand levels.

The RB-based cases are generally less affected by increased investment costs for non-commercial technologies, since only parts of the investment consist of non-commercial technologies. Removed support for green electricity influences the RB cases differently. However, the effect is in many cases positive, since the costs of wood fuel decrease significantly.

6.3.4.4 The influence of the possibility of CCS

If CCS is not an option, this limits the number of interesting cases for Scenarios 2 and 4, from both economic and environmental points of view, even to a larger extent than for Scenarios 1 and 3. However, for 10 GJ/ADt almost all the cases without CCS (8, 27-29) have a positive Δ NAP in Scenarios 2 and 4 (as well as in Scenarios 1 and 3), even in sensitivity analysis case *c*. All these cases also show a potential for reducing the CO_2 emissions. For 14.5 and 19 GJ/ADt, case 27 is the only case without CCS that shows a clear positive Δ NAP in Scenarios 2 and 4. However, especially in Scenario 2, NAP is reduced to being only slightly positive compared to the mill reference case if sensitivity analysis case *c* is considered. Case 27 increases global CO_2 emissions for these mill steam demand levels.

6.3.5 Conclusions

In this paper, black liquor gasification with downstream production of DME or electricity has been compared with other recovery boiler-based pulping biorefinery concepts from economic and climatic points of view. The economic performance and CO_2 emission balances have been calculated for different future energy market scenarios based on combinations of different fossil fuel price and CO_2 emission charge levels. Different types of mills, with different steam requirements, for which the applicability and performance of various biorefinery concepts vary, have been considered. The possibility to implement CCS technology in both combustion- and gasification-based systems has been included.

The following main conclusions can be drawn for future market pulp mills:

- Black liquor gasification with DME production has the best economic performance for all energy market scenarios considered, even if the policy incentive value for DME is reduced and the investment cost is significantly larger than reported in previous studies. The black liquor gasification plant should, from an economic point of view, be supplemented by a solid biomass gasification plant producing DME and electricity.

- Black liquor gasification with electricity production has fairly good economic performance in all scenarios, but not if the policy incentive value for green electricity is reduced and the investment cost is significantly larger than reported in previous studies.

- A recovery boiler-based investment including extraction of lignin has fairly good economic performance in scenarios based on a high fossil fuel price level and/or a high CO_2 charge.

- If there is a possibility for implementation of CCS in connection with the mill, it significantly improves the economic performance in scenarios based on a high CO_2 charge for both combustion- and gasification-based systems.

- All concepts that include CCS show a large potential for reduction of global CO_2 emissions. However, for the concepts without CCS, the CO_2 reduction potential is fairly moderate or low in scenarios based on a high CO_2 charge, and several cases lead to a net increase of global CO_2 emissions for scenarios based on a low CO_2 charge.

The following main conclusions can be drawn for future integrated pulp and paper mills:

- Black liquor gasification with DME production has the best economic performance in all scenarios. The plant should be supplemented by a bark boiler if only options with a reasonable need for external wood fuel are considered. This is valid even if the policy incentive value for DME is reduced and the investment cost is significantly larger than reported in previous studies. However, in this case the net annual profit in scenarios based on a low fossil fuel price level and a high CO_2 charge is low.

- The economic performance is poorer for almost all biorefinery options compared to for market pulp mills, which makes most options without CCS unprofitable. Extraction of lignin, for example, is only profitable in a scenario with a high fossil fuel price level in combination with a low CO_2 charge. In scenarios with a low CO_2 charge, a recovery boiler-based option integrated with solid biomass gasification with production of DME and electricity has a fairly good economic performance and could be interesting for integrated mills with a steam deficit.

- If there is a possibility for implementation of CCS, it significantly improves the economic performance in scenarios based on a high CO_2 charge for both combustion- and gasification-based systems, making almost all concepts profitable.

- Concepts that include CCS generally show a large potential for reduction of global CO_2 emissions. However, for the concepts without CCS, few options show potential for CO_2 reduction and most options lead to a significant net increase of global CO_2 emissions.

6.4 Paper VI

This section summarizes Paper VI, “Comparison of options for utilization of excess steam and debottlenecking the recovery boiler at kraft pulp mills – Systems analysis of economic performance and CO_2 emissions”. The aim of this paper was to compare different technologies, including black liquor gasification, for utilization of kraft pulp mill excess heat and debottlenecking the recovery boiler. Two different cases are considered: one where the pulp production capacity is unchanged and one where the pulp production capacity is increased by 25%. The studied technologies are compared with respect to NAP and global CO_2 emissions for different future energy market scenarios. A further analysis of how different parameters such as policy instruments and investment costs affect the different technologies is also included.

The studied energy system consists of a kraft market pulp mill (based on average Scandinavian technology) with the possibility to invest in energy efficiency measures (reducing the process steam demand) and new technologies. Investments in energy efficiency measures result in a steam surplus for the mill. The generated steam surplus enables the mill to increase its electricity production and/or produce other products. This, however, requires additional investments. The following main investment alternatives were considered:

- A BLGMF/DME plant with the possibility to capture CO_2
- A BLGCC plant with the possibility to capture CO_2
- A lignin extraction plant
- A back-pressure steam turbine and/or condensing steam turbine
- A CO_2 capture plant connected to the recovery boiler flue gases

There is also a possibility to invest in heat exchangers and/or heat pumps for delivery of excess heat to a district heating system. Two different cases are considered for pricing of the extracted lignin: one where lignin is priced as wood chips and one where it is priced as fuel oil. The possibility to import external wood fuel is not considered; the steam use has to be met by internal resources.

As described in Section 4.3, in this paper it is assumed that the recovery boiler is not technically in need of replacement. The minimum load of the recovery boiler is set to 55% of the nominal load. For both of the cases with black liquor gasification, the mill's steam balance is not the limiting factor for the maximum size of the plants; it is set by the minimum load of the recovery boiler. The pulp production capacity in this mill is 1000 ADt/day, as a contrast to the other papers where the capacity is 2000 ADt/day. Thus, the BLG plants in this paper have a capacity of about one fourth to one third (in the case with increased production capacity) of the capacity in the other papers and will consequently have significantly higher specific investment costs.

It is assumed that the recovery boiler already runs at maximum capacity, and consequently the mill has to either invest in a BLGMF plant, a BLGCC plant or a lignin extraction plant or upgrade the recovery boiler, in order to handle the increased amount of black liquor in the case of a production capacity increase.

As described in Section 5.2, this paper assumes a capital recovery factor of 0.2. This is considered a suitable choice since the investments are made in order to utilize a steam surplus. However, if a full substitution of the recovery boiler by a BLG plant is considered as in the other papers, a capital recovery factor closer to 0.1 is considered to be a more realistic alternative. The sensitivity analysis includes a case where the capital recovery factor is changed to 0.1.

When unchanged pulp production capacity is assumed, the availability of the BLG plant is not as critical as when the pulp production is increased or if a full substitution of the recovery boiler is considered, since the recovery boiler can handle the entire black liquor flow if necessary. Therefore, it is assumed that the "Nth" plant can appear earlier than if a full substitution is considered, and 2020 is also included to represent an average year upon which to base estimations of cash flows related to the different investment options. In this period it is assumed that infrastructure for CCS is not established, and therefore CCS is not retained as an investment option for the mill (or for technologies in the surrounding system).

For detailed descriptions of the studied system and cases, investment costs, scenarios and results, see Paper VI. From the results, the following conclusions can be drawn:

- Investment in energy efficiency measures and utilization of the resulting steam surplus for production of different energy products can significantly improve the economic performance of the pulp mill. Most of the studied cases significantly increase NAP compared to not investing in energy efficiency measures.

- In scenarios assuming a low fossil fuel price level, BLGMF generally achieves the best economic performance. In scenarios assuming a high fossil fuel price level, extraction of lignin that can be priced as oil achieves the best economic performance. The BLGMF case is, contrary to lignin priced as oil, very sensitive to changes of several parameters, especially the level of economic policy support for biofuels.

- All the studied technology cases decrease global CO_2 emissions significantly compared to not making investments. For the year 2020, when CCS is not assumed to be commercially available, BLGCC achieves the highest CO_2 reduction potential, followed by investments in new turbines in connection with the recovery boiler and extraction of lignin that can be priced as oil. For the year 2030, when there is assumed to be an established infrastructure for CCS, investments in CCS coupled to the recovery boiler flue gases yield the highest CO_2 reduction potential, followed by BLGCC and BLGMF, where CCS also can be included.

- The influence of the level of support for green electricity does not significantly influence the results, partly due to the assumed design of the support system where only new production capacity is entitled to support. However, the level of support for biofuels affects the results to a large extent, since it significantly influences the economic performance of the BLGMF case. If conditions influencing the BLGMF case positively are not considered, such as inclusion of CCS and increased pulp production capacity, a substantial level of the support for biofuels is needed in order for BLGMF to be competitive compared to extraction of lignin that can be priced as oil.

- The results show that for technologies with substantial investment costs, BLGMF, BLGCC and CCS coupled to the boiler flue gases, a 25% increase of the investment cost has a quite large influence on the economic performance. Since the investment costs for these non-commercial technologies are highly uncertain, this makes the future economic performance of these technologies hard to predict. For lignin extraction, which is not yet a commercialised technology but has a lower investment compared to the other non-commercial ones, a 25% increase of the investment cost has a very low influence on the economic performance.

- The possibility to capture CO_2 from the recovery boiler flue gases results in a large CO_2 reduction potential. However, the profitability of capturing the CO_2 is strongly dependent on the CO_2 charge. It is only for the scenarios with a high CO_2 charge that CCS coupled to the boiler flue gases is more profitable than investments in new turbines. CCS decreases the global CO_2 emissions and improves the economic performance for BLGMF and BLGCC both in absolute terms and in relation to the other technologies.

- BLGMF and BLGCC benefit from economy of scale and thus achieve a better economic performance when increasing the production capacity compared to when the production capacity remains unchanged. For the BLGMF case, this means that it is more profitable than extracting lignin that can be priced as oil for some of the scenarios, compared to when the production remains unchanged. For increased electricity production and CCS in connection with the recovery boiler, the production capacity increase affects the economic performance due to the fact that an additional investment in an upgrading of the recovery boiler *has* to be made.

- Extraction of lignin that can be priced as oil has a very good economic performance, even in the scenarios with a low fossil fuel price level. Furthermore, it is not highly influenced by any of the parameters studied in addition to the scenario parameters, and can therefore be said to be a fairly robust investment compared to BLGMF, which is highly influenced by several parameters such as biofuel policy instruments and investment costs. The CO_2 emissions reduction from lignin extraction is also fairly stable between the scenarios.

7 Discussion

Implementation of black liquor gasification and other pulping biorefinery concepts affects energy and material flows in the surrounding energy system. For example, DME from a BLGMF plant can replace fossil diesel and thereby decrease CO_2 emissions from the transport sector. However, such a BLGMF plant is a net importer of electricity, compared to a conventional recovery boiler powerhouse, thereby increasing CO_2 emissions in the power sector. In addition, the need for external wood fuel increases. BLGCC and lignin extraction technologies can be implemented in future market pulp mills without creating a steam deficit and making the mill dependent on external fuel. But when implemented in future integrated pulp and paper mills, the need for external wood fuel increases. For BLGMF concepts, all types of mills must import external wood fuel.

Biomass is a limited resource. In this thesis the biomass system is expanded to include alternative biomass use, by assuming that biomass used at the mill reduces the amount of biomass available for other applications in the surrounding energy system, thus increasing the CO_2 emissions from those applications.

There are large uncertainties concerning both the economic performance and CO_2 emission balances for future implementation of pulping biorefinery concepts, since the developments of energy prices, policy instruments and CO_2 emissions for marginal use of energy carriers are unknown. Thus, it is important to illustrate the influence of different possible future energy market conditions. In this thesis, energy market scenarios that reflect different future energy market conditions are considered. The scenarios are based on combinations of different fossil fuel price levels and CO_2 emission charge levels. The scenarios reflect the strong connection between different energy market parameters, and enable a packaged sensitivity analysis to be conducted.

For all energy market scenarios investigated in this thesis, BLGMF-based biorefinery concepts achieve the best economic performance. This conclusion remains valid even if the level of policy incentive support for biofuels is reduced and investment costs are significantly higher than reported in previous studies. The BLGMF plant considered in this work produces DME, retained as an example of a possible future biofuel. BLGMF-based concepts, as well as other biorefinery concepts, generally have better economic performance and lower CO_2 emissions for future market pulp mills compared to future integrated pulp and paper mills. This can be explained by the relatively low total efficiency for the recovery boiler-based market pulp mill reference investment option, due to power generation in a condensing steam turbine unit.

BLGCC-based biorefinery concepts achieve relatively good economic performance in all studied energy market scenarios for market pulp mills, but not if the level of policy incentive value for green electricity is reduced and the investment cost is significantly higher than reported in previous studies. For integrated mills, the profitability is very doubtful.

For future energy market conditions characterized by a low CO_2 emission charge, coal power plants are assumed to remain as marginal producers of electricity. Under these conditions, BLGCC contributes to reduction of global CO_2 emissions, especially for market pulp mills, whereas BLGMF leads to a significant net increase of global CO_2 emissions. In future scenarios with a high CO_2 emissions charge level, coal power plants equipped with CCS are assumed to become marginal producers of electricity. Under these conditions BLGCC only contributes to marginal reductions of global CO_2 emissions if implemented in market pulp mills, whereas global CO_2 emissions increase for implementation in integrated mills. BLGMF contributes to a significant decrease of CO_2 emissions for market pulp mills under these conditions, whereas for integrated mills the global CO_2 emissions increase.

An important assumption in this work is that a mill steam deficit is covered by firing bark and external wood fuel in a high-pressure steam boiler connected to a steam turbine. However, simultaneous implementation of solid biomass gasification technology with production of biofuels and/or electricity could also be considered. The results in this thesis show that this could improve the economic performance for BLGMF in market mills for all studied energy market scenarios, whereas for integrated mills the economic performance is only improved in scenarios based on a low CO_2 charge where the prices for biofuels and electricity are high in relation to the biomass feedstock price. Furthermore, for integrated mills the need for external wood fuel is very large if integration with solid biomass gasification is considered in addition to BLGMF. It can also be discussed whether it is realistic to consider investment options that require a mill to simultaneously implement several new technologies within a relatively short time frame. Thus, it might for example be more realistic to cover the steam deficit in a mill with a BLGMF plant with a conventional biomass CHP unit than with a solid biomass gasification plant producing motor fuels and/or electricity.

For future integrated pulp and paper mills with recovery boiler technology, integration of solid biomass gasification with production of biofuels and electricity has relatively good economic performance for energy market conditions characterized by a low CO_2 charge. However, this option increases global CO_2 emissions compared to covering the steam deficit with a conventional biomass CHP.

For market pulp mills, a recovery boiler-based biorefinery option including extraction of lignin has relatively good economic performance for energy market scenarios based on a high fossil fuel price level and/or high CO_2 charge, where the lignin price is medium or high. For integrated mills, however, the need for external wood fuel increases if lignin is extracted, and it is only profitable in scenarios with a high fossil fuel price level in combination with a low CO_2 charge, where the ratio between the lignin and biomass price is high. These conclusions are relatively insensitive to changes of the investment cost. It was assumed that the market value of lignin is determined by the willingness to pay for lignin as a substitute for oil feedstock for production of chemicals or materials. If implemented in market pulp mills, lignin extraction can contribute to reduction of global CO_2 emissions.

If CCS technology is commercially available and CO_2 collection infrastructure is available within the vicinity of the mill, its implementation can significantly improve the economic performance of both combustion- and gasification-based biorefinery concepts, for energy market scenarios with a high CO_2 charge. More CO_2 can be separated from the recovery boiler than from a BLGMF plant, but at a significantly higher cost. For a BLGCC plant, the cost for CCS is somewhat higher than for a BLGMF plant, but the separated amount of CO_2 is much higher and close to that for the recovery boiler. The indirect effects of CCS are, as discussed, also large. Implementation of CCS in the power sector significantly lowers the CO_2 reduction potential from processes with a surplus of renewable electricity such as BLGCC, whereas the reduction potential for processes with a net deficit of electricity such as BLGMF increases.

The potential CO_2 emissions reduction is generally large if CCS is implemented, in contrast to the relatively small, or non-existent, reduction potential shown by, for example, BLGMF processes operating without CCS. In BLGMF processes, where CO_2 is separated as part of the process, CCS appears to be profitable even in scenarios with a low CO_2 charge. However, it is doubtful whether there will be a widespread infrastructure for storing CO_2 if capture is not widely introduced in the power sector with its very large emission point sources. Furthermore, the majority of European kraft pulp mills are located in Scandinavia, far away from other large energy-intensive industries and potential fossil CO_2 capture clusters. Thus, even with a relatively widespread infrastructure for transportation and storage of CO_2 in central Europe, it is not at all certain that such infrastructure will be located within the vicinity of most kraft pulp mills.

Increased heat integration measures leading to steam savings should always be considered, especially if the mill has a steam deficit. In this thesis it has been shown that it is profitable for a future market pulp mill, both with recovery boiler and BLGMF technology, to invest in steam-saving measures. However, such measures are particularly profitable for the mill if BLGMF is implemented, especially in scenarios with a high CO_2 charge where the ratio between the biomass and electricity price is high. Decreasing the steam deficit means that the CHP plant can be made smaller, thus decreasing the need for external wood fuel, but also decreasing the internal electricity generation. This has a large effect on the incremental investment of investing in BLGMF plant instead of a new recovery boiler; the results indicate a significant decrease of the incremental investment cost. It has also been shown that increased heat integration decreases global CO_2 emissions. For future integrated mills, the effect of increased heat integration would be similar for all investment options, since it is likely that they will all have a steam deficit.

In order for mills to consider implementation of full-scale BLG plants, the recovery boiler must be close to the end of its technical lifetime. This has been an important assumption behind the results presented in this work. However, mills with a potential steam surplus or mills wishing to increase their production capacity (assuming that the recovery boiler already runs at maximum capacity) could consider investment in a smaller BLG plant as a way to utilize the potential steam surplus or to achieve debottlenecking of the recovery boiler. In such a case the BLG plant will be smaller, and it has also been assumed that the capital recovery factor will be higher than in the case of investment in a complete new plant for energy and chemical recovery. Thus, the specific capital cost will be significantly higher in these cases compared to investments in full-scale BLG plants. In these cases it has been shown that a number of conditions must occur simultaneously in order for BLGMF to be the most profitable option, compared for example to lignin extraction. This shows the importance of scale and choice of capital recovery factor. This thesis has generally assumed a relatively large mill size, from a Scandinavian perspective. This choice is based upon the trend in the pulp and paper industry towards fewer mills with larger capacity. The capital recovery factor is set at relatively low values typically associated with strategic core-business investments. However, in cases where a full substitution of the recovery boiler is not considered, a higher factor has been chosen.

The assumed level of policy instruments for biofuels was set based on the assumption that in order for Europe to achieve large-scale production of biofuels, the producers must be able to compete with other large-scale user categories for the biomass feedstock. It has been assumed that a potential marginal price-setting user category for biomass is coal power plants, benefiting from support for green electricity if co-firing biomass with coal. If stand-alone plants producing biofuels via gasification of solid biomass are to have the same willingness to pay for biomass under these conditions, a higher level of support for biofuels than for green electricity is necessary in scenarios with a low fossil fuel price level, whereas in scenarios with a high fossil fuel price level the necessary level of support for biofuels is lower than for green electricity. If implementation of CCS at the biofuel plant is not assumed to be possible, the levels of support required for biofuels needs to be very high in scenarios based on a low fossil fuel price level in combination with a high CO_2 charge. The CO_2 emissions reduction consequences of using biomass in a coal power plant are significantly higher than for biofuel production via gasification of solid biomass, if CCS is not considered. However, if CCS is considered, the CO_2 emissions reduction associated with biomass usage for biofuel production is close to the emissions reduction associated with biomass co-firing in coal power plants.

Projected commercial ("Nth plant") performance and costs are assumed in the calculations in this thesis. However, black liquor gasification still has to be successfully demonstrated in large scale in order to reach commercial status and constitute a viable alternative to the recovery boiler. This is also the case for the other non-commercial technologies, such as gasification of solid biomass. An important difference between black liquor gasification and the other non-commercial technologies considered in this thesis is that it is a part of the pulping process to a greater extent – required to continuously process pulping chemicals and provide the mill with green liquor.

8 Conclusions

This thesis has studied the influence of diverse factors on the economic performance and CO_2 emission balances associated with implementation of black liquor gasification and other pulping biorefinery concepts in future kraft pulp and paper mills. It has been assumed that if external wood fuel is used at the mill, this reduces the amount of wood fuel available for other applications, thereby increasing the CO_2 emissions from those applications. The following main conclusions can be drawn:

- Black liquor gasification and other pulping biorefinery concepts such as extraction of lignin generally have a better economic performance and more favourable CO_2 emission balances for future market pulp mills than for future integrated pulp and paper mills.

- Black liquor gasification with DME production proved to be the most profitable biorefinery concept investigated for all energy market scenarios considered. This conclusion holds even if the level of policy incentive support for DME is reduced and the plant investment costs are significantly larger than reported in previous studies.

- For market pulp mills, black liquor gasification with DME production could contribute to decreased global CO_2 emissions in scenarios where the marginal electricity production technology is coal power with CCS. For integrated pulp and paper mills, on the other hand, DME production from gasified black liquor leads to increased CO_2 emissions for all energy market scenarios considered.

- Simultaneous implementation of solid biomass gasification with production of DME and/or electricity could improve the profitability for black liquor gasification with DME production in market mills for all studied energy market scenarios. For integrated mills, profitability is only improved in scenarios with relatively high prices for DME and electricity in relation to wood fuel. Furthermore, for integrated mills the need for external wood fuel is very large.

- Black liquor gasification with electricity production has relatively good economic performance in all studied scenarios for market pulp mills, but not if the level of support for green electricity is reduced and the investment costs are significantly higher than reported in previous studies. For integrated mills, profitability is very doubtful.

- Black liquor gasification with electricity production leads to decreased CO_2 emissions if coal power plants without CCS are assumed as marginal producers of electricity. However, if CCS becomes commercially available for such plants, the CO_2 emissions reduction potential is very small for implementation of BLG biorefinery concepts in market pulp mills, and for integrated pulp and paper mills the global CO_2 emissions are in fact shown to increase.

- For market pulp mills, a recovery boiler-based biorefinery option including extraction of lignin for oil substitution has relatively good economic performance in scenarios where the lignin price is medium or high. For integrated mills, however, it is only profitable in scenarios where the ratio between the lignin and biomass price is high. This option is fairly insensitive to variation of the investment cost. If implemented in market pulp mills, lignin extraction can contribute to reduction of global CO_2 emissions.

- For integrated pulp and paper mills with recovery boiler technology, implementation of solid biomass gasification with production of biofuels and electricity has fairly good economic performance in scenarios with relatively high prices of DME and electricity in relation to biomass. However, this option increases global CO_2 emissions compared to investment in a conventional biomass CHP plant sized to cover the same steam demand.

- If implementation of CCS technology is an option for the mill, it can significantly improve profitability for both combustion- and gasification-based biorefinery concepts for energy market scenarios with a high CO_2 charge. Concepts that include CCS generally show a large potential for reduction of global CO_2 emissions.

- Increased heat integration measures leading to steam savings should always be considered, especially if the mill has a steam deficit. By considering increased heat integration, a significant reduction of the investment cost can be achieved and profitability can be significantly improved for biorefinery concepts such as black liquor gasification with DME production, especially in scenarios where the ratio between the biomass and electricity price is high. Furthermore, increased heat integration reduces global CO_2 emissions.

- Even if the recovery boiler is not close to the end of its technical lifetime, it could be interesting for mills to consider investment in a smaller BLG plant as a way to capitalize upon a possible steam surplus, or as a way to debottleneck the recovery boiler in combination with a production capacity increase.

9 Future work

The work presented in this thesis focused on the consequences of implementing biorefinery concepts based on black liquor gasification in different types of mills. Based on key data for kraft pulp and paper mills in Europe, such as production of black liquor, mill steam requirements and age structure of existing recovery boilers, the possible implementation rate of black liquor gasification in Europe can be estimated, as well as the consequences for the European energy system. Furthermore, with data of geographical location of the mills and the closeness to for example other industries, distribution infrastructure for biofuels, bulk chemicals, CO_2, etc., and the availability of biomass feedstock, the probability of different black liquor gasification-based biorefinery concepts can be estimated.

There is also need for more detailed studies of how to integrate black liquor gasification with different types of mills and, for example, with gasification of solid biomass feedstock. Production of motor fuels implies an increase of the number of new process streams and unit operations that is significantly greater than in the case of electricity generation. In addition, integration of black liquor gasification with electricity production has been studied in considerable detail in a number of previous studies. Therefore, the greatest need for future work is in the area of detailed process integration studies of black liquor gasification with downstream synthesis of different types of motor fuels in different kinds of mills.

There is an increasing level of research and development activities concerning extraction of lignin and hemicelluloses from the black liquor or pulp wood. Studies of these technologies in combination with black liquor gasification are needed to investigate how the black liquor gasification process is affected by modified black liquor compositions resulting from upstream lignin or hemicellulose extraction operations. As indicated in this work, the size of the gasification plant is important for economy of scale. Extraction of lignin and/or hemicelluloses will naturally significantly decrease the size of the black liquor gasification plant.

10 Nomenclature

Abbreviations

ADt	Air Dried tonne
BAT	Best Available Technology
BB	Bark Boiler
BFW	Boiler Feed Water
BIGCC	Biomass Integrated Gasification Combined Cycle
BIGMF	Biomass Integrated Gasification with Motor Fuel production
BLG	Black Liquor Gasification
BLGCC	Black Liquor Gasification Combined Cycle
BLGMF	Black Liquor Gasification with Motor Fuel production
BPEV	Battery Powered Electric Vehicle
CCS	Carbon Capture and Storage
CEPCI	Chemical Engineering Plant Cost Index
CHP	Combined Heat and Power
CP	Coal Power
CW	Cooling Water
DME	DiMethyl Ether
El	Electricity
FRAM	Future Resource Adapted pulp Mill
FT	Fisher-Tropsch
FTD	Fisher-Tropsch Diesel
GCC	Grand Composite Curve
GHG	GreenHouse Gases
GT	Gas Turbine
HP	High Pressure (steam)
HRSG	Heat Recovery Steam Generator
IHI	Increased Heat Integration
KAM	Eco-cyclic pulp mill (KretsloppsAnpassad Massafabrik)
LCA	Life Cycle Analysis
LE	Lignin Extraction
LLP	Low Low Pressure (steam)
LP	Low Pressure (steam)
MEA	Mono-EthanolAmines
MF	Motor Fuel
MP	Medium Pressure (steam)
NAP	Net Annual Profit
NGCC	Natural Gas Combined Cycle
O&M	Operation and Maintenance (cost)

RB	Recovery Boiler
ST	Steam Turbine
TRI	ThermoChem Recovery International
TTW	Tank-To-Wheel
WF	Wood Fuel
WTP	Willingness To Pay
WTT	Well-To-Tank
WTW	Well-To-Wheel

Chemical symbols

$Ca(OH)_2$	Calcium hydroxide
$CaCO_3$	Calcium carbonate
CaO	Calcium oxide
CH_4	Methane
CO	Carbon monoxide
CO_2	Carbon dioxide
COS	Carbonyl sulphide
H_2	Hydrogen
H_2O	Water
H_2S	Hydrogen sulphide
Na_2CO_3	Sodium carbonate
Na_2S	Sodium sulphide
NaOH	Sodium hydroxide

11 Acknowledgements

This project has been a part of the Energy Systems Programme, which is financed by the Swedish Energy Agency (SEA). Additional funding has been provided by the Black Liquor Gasification Program, which is financed by SEA, MISTRA, Chemrec, Smurfit Kappa, Södra Cell, SCA Packaging, Sveaskog and the County Administrative Board of Norrbotten, and also by the SEA's programme for Energy Efficiency in Industry.

There are a number of people whom I would like to thank, "some mentioned, none forgotten":

First of all I would like to thank my supervisor Simon Harvey for all discussions and guidance through this work and for always believing in me. Thank you for always making time for me in your busy schedule!

Thore Berntsson, thank you for interesting and valuable discussions and for sharing your thoughts about biomass usage, CCS, CO_2 consequences and so much more.

Anders Åsblad and Björn Sandén, thank you for valuable inputs to the work.

To Niklas Berglin, Ingvar Landälv, Erik Furusjö, Rikard Gebert, Jonas Rudberg and Lars Tegnér within the Black Liquor Gasification program, thank you for valuable inputs and discussions.

Elisabeth, my "speed-dial number one", thank you being such a good researcher and "boll-plank" during all these years. The cooperation with you has truly been great!

Mimmi, my "speed-dial number two", thank you for a pleasant cooperation and for all the nice talks during these years.

To all the D06:or within the Energy Systems Programme, thank you for such a nice year with courses. The discussion we had really broadened my perspectives! Special thanks to Magdalena and Erik for a nice cooperation in the Tvärprojekt.

Johanna, thank you for being such an excellent roommate and friend! Thank you for your encouragement and support during these years and for all the talks we have had. Thank you also for good research cooperation and valuable discussions.

To all my former and present colleagues, thank you for making my years at VoM so pleasant. Elin, thank you for our rewarding discussions and for friendship. Stefan, you were the perfect "ETM-partner". Mathias, thank you for always being so helpful. Rickard, thank you for including me in your project. Marcus, thank you for teaching me about OptiVap. Grégory, thank you for valuable assistance with simulations. Daniella, thank you for interesting discussions and for being such a good friend. Bengt, Raya, Birgitta, Lillemor and Nils, thank you for technical and administrative support. Lennart, thank you for your time answering questions about thermodynamics and energy technology – not just during the past five years but during my years as a student, it has really been valuable. And to all, thanks for fika, after work, luciatåg and numerous interesting discussions. It has really been a great time!

To my friends, thank you for supporting and believing in me and for all joyful times!

To my beloved family, Mamma, Pappa, Anna and Kristina, thank you for your unconditional love and support and for always being there for me.

Andreas, thank you for friendship and love. I am so happy that you are in my life!

12 References

Ahlvik P and Brandberg Å. Well-To-Wheel Efficiency for alternative fuels from natural gas or biomass. Publication 2001:85. Swedish National Road Administration, Borlänge, Sweden, 2001.

Ahlvik P and Brandberg Å. Sustainable fuels: Introduction of biofuels. Publication 2002:144. Swedish National Road Administration, Borlänge, Sweden, 2002.

Algehed J. Energy efficient evaporation in future kraft pulp mills. PhD Thesis. Department of Heat and Power Technology, Chalmers University of Technology, Göteborg, Sweden, 2002.

Andersson E. Benefits of Integrated Upgrading of Biofuels in Biorefineries – System Analysis. PhD Thesis. Department of Energy and Environment, Division of Heat and Power Technology, Chalmers University of Technology, Göteborg, Sweden, 2007.

Axelsson E. Energy Export Opportunities from Kraft Pulp and Paper Mills and Resulting Reductions in Global CO_2 Emissions. PhD Thesis. Department of Energy and Environment, Division of Heat and Power Technology, Chalmers University of Technology, Göteborg, Sweden, 2008.

Axelsson E and Harvey S. Scenarios for assessing profitability and carbon balances of energy investments in industry. AGS Pathways report 2010:EU1. AGS, The alliance for global sustainability. Pathways to sustainable European energy systems, Göteborg, Sweden, 2010.

Axelsson E, Olsson MR and Berntsson T. Increased capacity in kraft pulp mills: Lignin separation and reduced steam demand compared with recovery boiler upgrade. Nordic Pulp and Paper Research Journal 2006a;21(4):485-492.

Axelsson E, Olsson MR and Berntsson T. Heat integration opportunities in average Scandinavian kraft pulp mills: Pinch analyses of model mills. Nordic Pulp and Paper Research Journal 2006b;21(4):466-475.

Bergek A. Shaping and exploiting technological opportunities: The case of renewable energy technology in Sweden. PhD Thesis. Department of Industrial Dynamics, Chalmers University of Technology, Göteborg, Sweden, 2002.

Berglin N. Pulp mill energy systems with black liquor gasification: a process integration study. Licentiate Thesis. Deptartment of Heat and Power Technology, Chalmers University of Technology, Göteborg, Sweden, 1996.

Berglin N and Andersson L. Process Integration of Black Liquor Gasification for Incremental Recovery Capacity. In Proceedings of International Chemical Recovery Conference, Whistler, British Columbia, Canada, June 11-14, 2001.

Berglin N and Berntsson T. CHP in the pulp industry using black liquor gasification: thermodynamic analysis. Applied Thermal Engineering 1998;18(11):947-961.

Berglin N, Eriksson H and Berntsson T. Performance Evaluation of Competing Designs for Efficient Cogeneration from Black Liquor. 2nd Biennial Johan Gullichsen Colloquium, Helsinki, Finland, September 9-10, 1999.

Berglin N, Lindblom M and Ekbom T. Preliminary economics of black liquor gasification with motor fuels production. In Proceedings of Colloquium on Black Liquor Combustion and Gasification, Park City, Utah, USA, May 13-16, 2003.

Berglin N, Tomani P, Olowsson P, Hultberg T and Persson S. Expersiences from Feeeding and Co-firing of Lignin Powder in a Lime Kiln. In Proceedings of International Chemical Recovery Conference, Williamsburg, Virginia, USA, 29 March – 1 April, 2010;1:175-187.

Berndes G, Hoogwijk M and van den Broek R. The contribution of biomass in the future global energy supply: a review of 17 studies. Biomass & Bioenergy 2003;25(1):1-28.

Biermann CJ. Handbook of Pulping and Papermaking (Second Edition). Academic Press, San Diego, 1996. *Available at: http://www.sciencedirect.com/science/book/9780120973620.*

Brau JF. Energy efficiency measures in an average Swedish Kraft pulp mill with hemicelluloses pre-extraction, Near-neutral and Alkaline extraction. Master Thesis. Department of Energy and Environment, Division of Heat and Power Technology, Chalmers University of Technology, Göteborg, Sweden, 2010.

Börjesson P. Good or bad bioethanol from a greenhouse gas perspective – What determines this?. Applied Energy 2009;86(5):589-594.

CEPI. Annual Statistics 2007 – European Pulp and Paper Industry. Confederation of European Paper Industries, 2008.

CEPI. Key Statistics 2008 – European Pulp and Paper Industry. Confederation of European Paper Industries, 2009.

Chemrec. Process for energy and chemicals recovery from black liquor in pulp mills, Booster. 2005a. *Available at: www.chemrec.se.*

Chemrec. Process for energy and chemicals recovery from black liquor in pulp mills, DP-1. 2005b. *Available at: www.chemrec.se.*

Delin L, Berglin N, Eriksson T, Andersson R, Samuelsson Å, Lundström A, Backlund B, Sivard Å and Åberg M. Integrated fine paper mill, FRAM 10. STFI-Packforsk, Stockholm, Sweden, 2005a.

Delin L, Berglin N, Lundström A, Samuelsson Å, Backlund B and Sivard Å. Bleached market kraft pulp mill, Reference and type mill, FRAM 09. STFI-Packforsk, Stockholm, Sweden, 2005b.

Delucchi M. Lifecycle analyses of biofuels. Draft report. Publication no. UCD-ITS-RR-06-08. Institute of Transportation Studies, University of California, Davis, 2006.

Difs K, Wetterlund E, Trygg L and Söderström M. Biomass gasification opportunities in a district heating system. Biomass and Bioenergy 2010;34(5):637-651.

Edwards R, Larivé J-F, Mahieu V and Rouveirolles P. Well-to-wheels analysis of future automotive fuels and powertrains in the European context, version 2c. JRC, EUCAR and CONCAWE, 2007.

Ekbom T, Berglin N and Lögdberg S. Black Liquor Gasification with Motor Fuel Production – BLGMF II. Nykomb Synergetics, Stockholm, Sweden, 2005.

Ekbom T, Lindblom M and Berglin N. Technical and commercial feasibility study of black liquor gasification with methanol/DME production as motor fuels for automotive uses. Alterner II report, 2003.

Ekvall T. Key methodological issues for life cycle inventory analysis of paper recycling. Journal of Cleaner Production 1999;7(4):281-294.

Ekvall T and Weidema BP. System boundaries and input data in consequential life cycle inventory analysis. International Journal of Life Cycle Assessment 2004;9(3):161-171.

Eriksson H. System aspects of black liquor gasification: consequences for both industry and society. Licentiate Thesis. Department of Heat & Power Technology, Chalmers University of Technology, Göteborg, Sweden, 2001.

ETC. Gasification of black liquor. Popular science report from the BLG II program 2007-2010. 2011. *Available at: http://www.etcpitea.se/en/nyheter/popular-science-report-from-the-blg-ii-program/.*

European Commission. Communication from the Commission – An EU strategy for biofuels. COM(2006) 34. 2006.

European Commission. Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and Committee of the Regions. Energy 2020 – A strategy for competitive, sustainable and secure energy. 2010.

European Union. Directive 2009/28/EC of the European Parliament and of the Council of 23 April 2009 on the promotion of the use of energy from renewable sources and amending and subsequently repealing Directives 2001/77/EC and 2003/30/EC. 2009.

Finnveden G, Hauschild MZ, Ekvall T, Guinée J, Heijungs R, Hellweg S, Koehler A, Pennington D and Suh S. Recent developments in Life Cycle Assessment. Journal of Environmental Management 2009;91(1):1-21.

Fleming JS, Habibi S and MacLean HL. Investigating the sustainability of lignocellulose-derived fuels for light-duty vehicles. Transportation Research Part D – Transport and Environment 2006;11(2):146-159.

Fornell R, Pettersson K and Berntsson T. Preliminary Design and Energy Efficiency Analysis of a Kraft Pulp Mill Converted to a Biorefinery Producing Ethanol and DME from Softwood. PRES 2010, 13th International Conference on Process Integration, Modelling and Optimisation for Energy Saving and Pollution Reduction, Chemical Engineering Transactions 2010;21:1147-1152.

FRAM. FRAM Final report Application area: Model mills and system analysis, FRAM Report No 70. STFI-Packforsk, Stockholm, Sweden, 2005.

Furusjö E. Process Engineer, Chemrec AB. Personal Communication, 2011-03-18.

Gustavsson L, Holmberg J, Dornburg V, Sathre R, Eggers T, Mahapatra K and Marland G. Using biomass for climate change mitigation and oil use reduction. Energy Policy 2007;35(11):5671-5691.

Gustavsson L and Karlsson Å. CO_2 mitigation: On methods and parameters for comparison of fossil-fuel and biofuel systems. Mitigation and Adaptation Strategies for Global Change 2006;11(5-6):935-959.

Hamelinck CN and Faaij APC. Outlook for advanced biofuels. Energy Policy 2006;34(17):3268-3283.

Harvey S and Facchini B. Predicting black liquor gasification combined cycle powerhouse performance accounting for off-design gas turbine operation. Applied Thermal Engineering 2004;24(1):111-126.

Hektor E. Post-combustion CO_2 capture in kraft pulp and paper mills – Technical, economic and systems aspects. PhD Thesis. Department of Energy and Environment, Division of Heat and Power Technology, Chalmers University of Technology, Göteborg, Sweden, 2008.

Hellsmark H. Unfolding the formative phase of gasified biomass in the European Union – The role of the system builders in realising the potential of second-generation transportation fuels from biomass. PhD Thesis. Department of Energy and Environment, Division of Environmental Systems Analysis, Chalmers University of Technology, Göteborg, Sweden, 2010.

Hillman K. Envioronmental Assessment and Strategic Technology Choice – The Case of Renewable Transport Fuels. PhD Thesis. Department of Energy and Environment, Division of Environmental Systems Analysis, Chalmers University of Technology, Göteborg, Sweden, 2008.

Hillman KM and Sanden BA. Time and scale in Life Cycle Assessment: The case of fuel choice in the transport sector. International Journal of Alternative Propulsion 2008;2(1):1-12.

Holmgren K, Eriksson E, Olsson O, Olsson M, Hillring B and Parikka M. Biofuels and climate neutrality – system analysis of production and utilisation. Elforsk, Stockholm, Sweden, 2007.

Hoogwijk M. On the global and regional potential of renewable energy sources. PhD Thesis. Utrecht University, Utrecht, the Netherlands, 2004.

IPCC. Fourth assessment report. Climate change 2007: Synthesis report. Summary for policymakers. 2007. *Available at: http://www.ipcc.ch/publications_and_data/ar4/syr/en/spm.html.*

Isaksson A. Flexible and Efficient Biomass Energy Systems in the Pulp Industry. Licentiate Thesis. Department of Chemical Engineering and Technology, Energy Processes, Royal Institute of Technology, Stockholm, Sweden, 2000.

ISO. Environmental Management – Life cycle assessment – Requirements and guidelines (ISO 14044:2006). European Committee for Standardization, 2006.

Joelsson JM and Gustavsson L. CO_2 emission and oil use reduction through black liquor gasification and energy efficiency in pulp and paper industry. Resources, Conservation and Recycling 2008;52(5):747-763.

Joelsson JM, Gustavsson L, Pingoud K and Soimakallio S. CO_2 balance and oil use reduction of syngas-derived motor fuels co-produced in pulp and paper mills 17th European Biomass Conference & Exhibition, Hamburg, Germany, 29 June – 3 July, 2009.

Järås S. Professor, Royal Institute of Technology. Personal Communication, 2006-12-07.

Jönsson J and Algehed J. Pathways to a sustainable European kraft pulp industry: Trade-offs between economy and CO_2 emissions for different technologies and system solutions. Applied Thermal Engineering 2010;30(16):2315-2325.

Jönsson J and Berntsson T. Analysing the Potential for CCS within the European Pulp and Paper Industry. In Proceedings of 23rd International Conference on Efficiency, Cost, Optimization, Simulation and Environmental Impact of Energy Systems (ECOS), Lausanne, Switzerland, June 14-17, 2010;676-683.

Jönsson J, Svensson I-L, Berntsson T and Moshfegh B. Excess heat from kraft pulp mills: Trade-offs between internal and external use in the case of Sweden – Part 2: Results for future energy market scenarios. Energy Policy 2008;36(11):4186-4197.

KAM. KAM-report A100, Final report from the Ecocyclic pulp mill programme. STFI, Stockholm, Sweden, 2003.

Kartha S, Lazarus M and Bosi M. Baseline recommendations for greenhouse gas mitigation projects in the electric power sector. Energy Policy 2004;32(4):545-566.

Landälv I. Technical Director, Chemrec AB. Personal communication, 2007-04-20.

Landälv I, Granberg F and Nelving H. Production of Di Methyl Ether from Kraft Black Liquor for Use in Heavy Duty Trucks. In Proceedings of International Chemical Recovery Conference, Williamsburg, Virginia, USA, 29 March - 1 April, 2010;2:179-186.

Larson E. A review of life-cycle analysis studies on liquid biofuel systems for the transport sector. Energy for Sustainable Development 2006;X(2):109-126.

Larson ED, Consonni S and Katofsky RE. A Cost-Benefit Assessment of Biomass Gasification Power Generation in the Pulp and Paper Industry. 2003. *Available at: www.princeton.edu/~energy.*

Larson ED, Consonni S, Katofsky RE, Iisa K and Frederick JW. A Cost-Benefit Assessment of Gasification-Based Biorefining in the Kraft Pulp and Paper Industry. Final report (in four volumes). 2006. *Available at: www.princeton.edu/~energy.*

Larson ED, Consonni S, Katofsky RE, Iisa K and Frederick JW. An assessment of gasification-based biorefining at kraft pulp and paper mills in the United States, Part A: Background and assumptions. Tappi Journal 2008;November:8-14.

Larson ED, Consonni S, Katofsky RE, Iisa K and Frederick JW. An assessment of gasification-based biorefining at kraft pulp and paper mills in the United States, Part B: Results. Tappi Journal 2009;January:27-35.

Lindfeldt EG, Saxe M, Magnusson M and Mohseni F. Strategies for a road transport system based on renewable resources – The case of an import-independent Sweden in 2025. Applied Energy 2010;87(6):1836-1845.

Lindström M, Jameel H, Naitbani V, Kirkman A and Renard J. Integrating black liquor gasification and pulping: A review of current technology. ACS Symposium Series 2007;954:434-462.

Linnhoff B. Pinch analysis – A state-of-the-art overview. Chemical Engineering Research and Design 1993;71(A5):503-522.

MacLean HL and Lave LB. Evaluating automobile fuel/propulsion system technologies. Progress in Energy and Combustion Science 2003;29(1):1-69.

Marbe Å. New Opportunities and System Consequences for Biomass Integrated Gasification Technology in CHP Applications. PhD Thesis. Department of Energy and Environment, Division of Heat and Power Technology, Chalmers University of Technology, Göteborg, Sweden, 2005.

Maunsbach K, Isaksson A, Yan J, Svedberg G and Eidensten L. Integration of advanced gas turbines in pulp and paper mills for increased power generation. Journal of Engineering for Gas Turbines and Power-Transactions of the ASME 2001;123(4):734-740.

McKeough P and Kurkela E. Process evaluations and designs in the UCG project 2004-2007. VTT, Espoo, Finland, 2008.

Merrild H, Darngaard A and Christensen TH. Life cycle assessment of waste paper management: The importance of technology data and system boundaries in assessing recycling and incineration. Resources Conservation and Recycling 2008;52(12):1391-1398.

Modig G. Black liquor gasification: an assessment from the perspective of the pulp and paper industry. Licentiate Thesis. Environmental and Energy Systems Studies, Lund University, Lund, Sweden, 2005.

Möllersten K. Opportunities for CO_2 reduscions and CO_2-lean energy systems in pulp and paper mills. PhD Thesis. Department of Chemical Engineering and Technology, Division of Energy Processes, Royal Institute of Technology, Stockholm, Sweden, 2002.

Nyström I, Ahlgren E, Andersson E, Börjesson M, Fahlén E, Harvey S, Ingman D, Karlsson S, Larsson E and Löfgren Å. Biokombi Rya, biobränsleförgasning satt i system. Slutrapport från forskningsprojektet Biokombi Rya. CEC, Chalmers EnergiCentrum, rapport CEC 2007:2. 2007. *(In Swedish)*

Näsholm A-S and Westermark M. Energy studies of different cogeneration systems for black liquor gasification. Energy Conversion and Management 1997;38(15-17):1655-1663.

Olsson MR. Simulations of Evaporation Plants Including Lignin Extraction and Using Excess Heat in Kraft Pulp Mills. PhD Thesis. Department of Energy and Environment, Division of Heat and Power Technology, Chalmers University of Technology, Göteborg, Sweden, 2009.

Olsson MR, Axelsson E and Berntsson T. Exporting lignin or power from heat-integrated kraft pulp mills: A technoeconomic comparison using model mills. Nordic Pulp and Paper Research Journal 2006;21(4):476-484.

Renew. Renewable Fuels for advanced Powertrains. Final Report. 2008. *Available at: http://www.renew-fuel.com.*

Rudberg J. Managing Director, Chemrec AB. Personal communication, 2006-11-24.

Schlamadinger B, Apps M, Bohlin F, Gustavsson L, Jungmeier G, Marland G, Pingoud K and Savolainen I. Towards a standard methodology for greenhouse gas balances of bioenergy systems in comparison with fossil energy systems. Biomass & Bioenergy 1997;13(6):359-375.

Schlamadinger B, Edwards R, Byrne K, Cowie A, Faaij A, Green C, Fijan-Parlov S, Gustavsson L, Hatton T, Heding N, Kwant K, Pingoud K, Ringer M, Robertson K, Solberg B, Soimakallio S and Woess-Gallasch S. Optimizing the greenhouse gas benefits of bioenergy systems. In Proceedings of 14th European Biomass Conference, Paris, France, October 17-21, 2005;2029-2032.

SEA. The Swedish Energy Agency, http://www.energimyndigheten.se/sv/Press/Pressmeddelanden/EU-kommissionen-godkanner-statligt-stod-for-att-utveckla-andra-generationens-biodrivmedel/, 2011-02-28.

Stevens DJ. Hot gas conditioning: Recent progress with larger-scale biomass gasification systems. Update and summary of recent progress. NREL/SR-510-29952. National Renewable Energy Laboratory, Colorado, USA, 2001.

Swedish Forest Industries. Skogsindustriernas miljödatabas: Bruk 2009. 2009.

Tegnér L. Swedish Energy Agency. Personal Communication, 2007-03-22.

Tillman A-M. Significance of decision-making for LCA methodology. Environmental Impact Assessment Review 2000;20(1):113-123.

TRI. ThermoChem Recovery International, http://www.tri-inc.net/, 2011-03-23.

Weidema BP, Frees N and Nielsen AM. Marginal production technologies for life cycle inventories. International Journal of Life Cycle Assessment 1999;4(1):48-56.

Wetterlund E, Pettersson K and Harvey S. Systems analysis of integrating biomass gasification with pulp and paper production – Effects on economic performance, CO_2 emissions and energy use. Energy 2011;36(2):932-941.

Whitty K and Nilsson A. Experience from a high temperature, pressurized black liquor gasification pilot plant. In Proceedings of International Chemical Recovery Conference, Whistler, British Columbia, Canada, 2001;281-287.

Whitty K and Verrill CL. A historical look at the development of alternative black liquor recovery technologies and the evolution of black liquor gasifier designs. In Proceedings of International Chemical Recovery Conference, Charleston, SC, USA, 2004;1:13-33.

Wising U, Berntsson T and Åsblad A. Usable excess heat in future kraft pulp mills. Tappi Journal 2002;November:27-29.

Ådahl A and Harvey S. Energy efficiency investments in Kraft pulp mills given uncertain climate policy. International Journal of Energy Research 2007;31(5):486-505.